

Scattering of light by electrons in nonuniform semiconductor alloys

F. T. Vas'ko and Yu. N. Soldatenko

Institute of Semiconductor Physics, Ukraine Academy of Sciences, 252650 Kiev, Ukraine

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We calculate the contribution to the cross section for light scattering connected with spatial nonuniformity of the electron inverse effective mass in semiconductor alloys. When the distance l_c over which the effective mass varies exceeds the screening radius, the nonuniformities are unscreened, and their contribution can exceed those of scattering mechanisms that operate in uniform materials. We obtain the spectral dependence for the cross section in the collisionless and hydrodynamic scattering regimes; the transition between these regimes occurs when l_c is on the order of the mean free path.

1. INTRODUCTION

The electron scattering of light in strongly doped semiconductor materials has been investigated in a number of papers (see the review Refs. 1–3). The usual mechanism for light scattering by charge density fluctuations (where the total scattering cross section is given by the Thomson cross section multiplied by the mean-square concentration fluctuations⁴) is found to be unimportant in these materials, because the contribution of such fluctuations to the scattering cross section is effectively screened. This scattering is suppressed by the presence of the small factor $(r_{sc}q)^4$ in the structure factor $S(\omega, \mathbf{q})$ (where r_{sc} is the screening radius, \mathbf{q} is the momentum transfer, and ω is the change in frequency of the scattered radiation). Appreciable scattering by free carriers can only come from contributions to the cross section from unscreened fluctuations in the parameters that determine the semiconductor band structure, e.g., effective-mass nonparabolicity or anisotropy (in a multivalley semiconductor), fluctuation of the g -factor in a magnetic field, or the quadrupole moment of holes in p -type material.

The situation changes in strongly doped semiconductor alloys with spatially nonuniform composition (the causes of such nonuniformities can be technology-related). If the length scale over which the composition varies, as defined by a certain correlation length l_c , exceeds r_{sc} , small long-wavelength nonuniformities of the potential energy at the conduction band extremum will be screened (the material will be considered n -type).¹ The effective mass of an electron also changes with the alloy composition; however, nonuniformities in the kinetic energy are screened only “on the average.” For this reason, the dispersion law E_{pr} for electrons in strongly doped alloys was given in Ref. 6 by the expression

$$E_{pr} = \varepsilon_p + \alpha \delta_r (\varepsilon_F - \varepsilon_p), \quad (1)$$

where $\varepsilon_p = p^2/2m$, $m = \langle m_r \rangle$; we have introduced the rate of change of the effective mass with composition α via by the relation $\alpha \delta_r = (m_r - m)/m$. The quantity $\delta_r = x_r - \langle x_r \rangle$ describes variations in the composition for substitutional alloys of the form $A_x \bar{A}_{1-x} B$; $\langle \dots \rangle$ denotes averaging over these nonuniformities; and ε_F is the Fermi energy. Alloys

can thus exhibit a new type of disorder, associated with nonuniformity of the kinetic rather than the potential energy of a charge carrier. The unscreened variations in the effective mass appearing in (1) lead to an additional mechanism for light scattering. Since the Thomson cross section is determined by the classical electron radius $r_e = e^2/mc^2$, for a nonuniform semiconductor alloy in which $r_{sc} < l_c < \bar{\delta}/q$ holds (where $\bar{\delta}$ is the mean-square deviation of the alloy composition from $\langle x_r \rangle$) the structure factor contains the quantity $\bar{\delta}^2 (r_{sc}/l_c)^4$, which replaces the small factor $(r_{sc}q)^4$ mentioned above. Even when the nonuniformity of the alloy is small, this contribution can exceed the contributions from unscreened scattering mechanisms.^{1–3} We also note that when this light scattering mechanism operates, the condition for a transition between the regimes of collisionless and diffuse scattering also changes [these regimes corresponds to Gaussian and Lorentzian spectral functions $S(\omega, \mathbf{q})$]. Now this transition takes place for values of l_c on the order of the mean free path l_F , and not for $ql_F \sim 1$, as in the case of scattering by charge fluctuations or band-structure parameters in a uniform material.

In Sec. 2, which follows, we will derive an expression for the scattering cross section by unscreened nonuniformities in the electron effective mass; in Sec. 3 we compute the structure factor that determines the spectrum for light scattering. In the Conclusion we compare this mechanism with other scattering mechanisms.

2. CONTRIBUTION OF EFFECTIVE-MASS VARIATIONS TO THE SCATTERING CROSS SECTION

The scattering cross section is determined by contributions to the alloy Hamiltonian in the effective-mass approximation that are quadratic in the vector potential:⁸

$$\left(\frac{e}{c}\right)^2 \sum_j \mathbf{A}(\mathbf{r}_j)^2 / 2m(\mathbf{r}_j), \quad (2)$$

(here j labels the electrons) and that contain the unscreened effective-mass nonuniformities introduced in (1). When these nonuniformities are taken into account, the

expression for the differential scattering cross section can be derived according to the general schemes of Refs. 2 and 9, leading to the expression

$$\left(\frac{d^2\sigma}{d\omega_s d\Omega} \right)_\mu = r_e^2 (\mathbf{e}_I \mathbf{e}_S)^2 \frac{\omega_s}{\omega_I} S(\omega, \mathbf{q}) \quad (3)$$

in which we have factored out $(\mathbf{e}_I \mathbf{e}_S)^2$ for the polarization dependence (this dependence does not change when we include the unscreened effective-mass nonuniformities; \mathbf{e}_I and \mathbf{e}_S are polarization unit vectors, and ω_I and ω_s are the frequencies of the incident and scattered radiation) and the structure factor

$$S(\omega, \mathbf{q}) = \sum_{i,f} F_i \left| \left(f \left| \sum_j \Phi(\mathbf{r}_j) \right| i \right) \right|^2 \delta(\varepsilon_i - \varepsilon_f + \hbar\omega). \quad (4)$$

Here i and f enumerate the initial and final states of the many-electron system with energies ε_i and ε_f ; F_i is the equilibrium distribution of electrons; and we will assume that $\hbar\omega_{1,s}$ are large compared to the Fermi energy ε_F . In (3), (4) we have not included the variation in the alloy dielectric constant, i.e., the incident and scattered modes are assumed to be plane waves; in addition, the short-wavelength components of the electromagnetic field are discarded. This approximation is a fairly good description in $\text{Al}_x\text{Ga}_{1-x}\text{As}$, in which $\alpha \simeq 1.25$; the rate of change of the dielectric permittivity with composition as defined by analogy with α is 5 times smaller.

The contribution from unscreened effective-mass nonuniformities is described by the matrix element in (4), in which the density fluctuation proportional to $\exp(i\mathbf{q}\mathbf{r})$ is replaced by the following factor:

$$\Phi(\mathbf{r}) = \exp(i\mathbf{q}\mathbf{r}) \left(\frac{m}{m(\mathbf{r})} \right). \quad (5)$$

In calculating the scattering cross section, we use the usual relation (whose derivation was given in Refs. 10 and 2) between the structure factor and the response to an external field proportional to $\Phi(\mathbf{r})\exp(-i\omega t)$. As a result, the structure factor defined by the right side of (4) is given by the expression

$$S(\omega, \mathbf{q}) = - \frac{\hbar \langle \int d\mathbf{r} \Phi(\mathbf{r}) * \Delta\rho(\mathbf{r}) \rangle}{4\pi e^2 [1 - \exp(-\hbar\omega/T)]}, \quad (6)$$

where the absorbed power averaged over fluctuations is here specified by the induced charge density $\Delta\rho(\mathbf{r}) \times \exp(-i\omega t)$, which is calculated in the self-consistent approximation. The relation between $\Delta\rho(\mathbf{r})$ and $\Phi(\mathbf{r})$ is found from the Poisson equation and the kinetic equation describing the linear response of the electrons to the perturbation (5) (which varies on the spatial and temporal scales $\min(l_c, q^{-1})$ and ω^{-1} , respectively). We can write the kinetic equation for a nonuniform semiconductor alloy and pass to the hydrodynamic approximation by analogy with Ref. 11. However, in calculating (4) up to accuracy of order $\bar{\delta}^2$, we may neglect fluctuations in the composition in writing the relation between the induced charge and the scattering potential, and ignore the effective-mass varia-

tions in the kinetic equation.²⁾ In this case the power absorbed in (6) is expressed in terms of the permittivity $\varepsilon(\omega, \mathbf{q})$ through the following relation (where $\bar{\kappa}$ is the permittivity of the lattice, whose dispersion is unimportant):

$$S(\omega, \mathbf{q}) = - \frac{\bar{\kappa}}{(4\pi e)^2} \frac{\hbar\omega}{[1 - \exp(-\hbar\omega/T)]} \frac{1}{V} \times \sum_k k^2 \langle \Phi(\mathbf{k}) * \Phi(\mathbf{k}) \rangle \text{Im} \frac{\bar{\kappa}}{\varepsilon(\omega, \mathbf{k})}, \quad (7)$$

$$\langle \Phi(\mathbf{k}) * \Phi(\mathbf{k}) \rangle = V [V\delta_{\mathbf{k},\mathbf{q}} + \alpha^2 W(|\mathbf{k} - \mathbf{q}|)]. \quad (8)$$

These expressions generalize the usual result for scattering by fluctuations in the charge density, because (7) contains the average (8) in which the second term defines the contribution to the cross section from unscreened effective-mass nonuniformities; $W(k)$ is a Gaussian correlator that describes the compositional nonuniformity, and V is the normalization volume.

3. SPECTRAL DEPENDENCE OF THE STRUCTURE FACTOR

Let us now consider how the contribution of the last term in (8) changes the structure factor (4). Using the collisionless approximation (where the mean free path is large compared to the scale of the nonuniformities, i.e., $l_F \gg l_c$), we obtain the following expression from (7) and (8):

$$S(\omega, \mathbf{q}) = \frac{3}{4} \frac{nV\hbar/\varepsilon_F}{[1 - \exp(-\hbar\omega/T)]} \left[(r_{sc}q)^4 F\left(\frac{\omega}{qv_F}\right) + (\alpha\bar{\delta})^2 \left(\frac{r_{sc}}{l_c}\right)^4 G\left(\frac{\omega}{v_F l_c}, \frac{ql_c}{2}\right) \right]. \quad (9)$$

Here n is the concentration of strongly degenerate electrons; the spectral dependence of the small contribution from density fluctuations (i.e., the first term of (9) proportional to $(r_{sc}q)^4$) is given by the function

$$F(\bar{y}) = \frac{\bar{y}\Theta(1 - |\bar{y}|)}{\left(1 - \bar{y} \ln \left| \frac{1 + \bar{y}}{1 - \bar{y}} \right| \right)^2 + (\pi\bar{y}/2)^2}, \quad (10)$$

which is obtained by using $\varepsilon(\omega, \mathbf{q})$ from Ref. 10. The spectral dependence of the contribution of effective-mass fluctuations to the scattering cross section is given by the second term of (9), in which the function G is defined by the relation

$$\bar{\delta}^2 G\left(\frac{\omega}{v_F l_c}, \frac{ql_c}{2}\right) = \frac{1}{V} \sum_k W(|\mathbf{k} + \mathbf{q}|) (kl_c)^4 F\left(\frac{\omega}{kv_F}\right). \quad (11)$$

This function increases linearly with frequency for small ω , and falls off with exponential sharpness at high frequencies. Figure 1 shows a plot of the spectral functions for several values of $(ql_c/2)$; the functions F and G are odd in ω , so that $S(\omega, \mathbf{q}) > 0$. Thus, scattering by effective-mass fluctuations leads to a broad spectral line independent of the scattering wave vector, whose intensity, according to (9)–

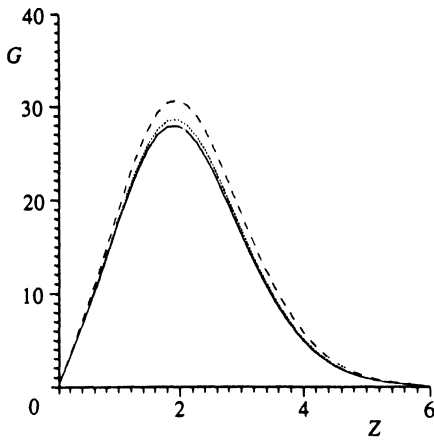


FIG. 1. Scattering cross section spectral functions [see (9) and (11)] $G[Z, ql_c/2]$ for $ql_c/2=0$ (solid curve), 0.1 (dots), and 0.2 (dashed curve).

(11), exceeds the intensity of single-particle scattering by charge density fluctuations when the condition $\bar{\delta}\alpha > ql_c$ holds.

In the hydrodynamic limit (i.e., when q^{-1} and l_c are small compared to l_F), we can use the fact that $\text{Im } \varepsilon(\omega, \mathbf{q})^{-1}$ is independent of wave vector, so that the factor $\sum_{\mathbf{k}} [k^2 \langle \Phi(\mathbf{k})^* \Phi(\mathbf{k}) \rangle]$ enters into $S(\omega, \mathbf{q})$. As a result of integrating, we obtain

$$S(\omega, \mathbf{q}) \sim [q^2 + 6(\alpha\bar{\delta}/l_c)^2], \quad (12)$$

so that the contributions to the structure factor due to charge density fluctuations (the first term) and effective-mass fluctuations (the second term) give the same spectral functions. These contributions are qualitatively different only in their dependence on the momentum transfer q : scattering by density fluctuations depends on the geometry of the scatterer, whereas the contribution from effective-mass nonuniformities does not. The latter mechanism will dominate only in the case of fine-scale fluctuations, when the condition $\bar{\delta}^2 \gg (ql_c)^2/6\alpha^2$ holds.

An intermediate case is possible, i.e., $q^{-1} > l_F > l_c$, where the scattering by density fluctuations is described using the hydrodynamic approximation, while the effective-mass nonuniformities are described using the collisionless regime. In this case, the expression for $S(\omega, \mathbf{q})$ is obtained by a combination of the expressions given above.

4. CONCLUSION

Let us now pause to discuss the conditions for observing this scattering mechanism by comparing the cross sections determined by (9) and (11), i.e., in the collisionless scattering regime, with the analogous expressions for scattering by energy density fluctuations (i.e., where the effective mass varies due to nonparabolicity of the energy spectrum; see Refs. 12 and 2).³⁾ By comparing the maximum values of the structure factor for this mechanism (assuming that $\hbar\omega_{1s}$ are smaller than the width of the forbidden band ε_G , and ignoring resonant enhancement of the scattering) with those determined by (9), we obtain the condition

$$(\delta\alpha)^2 \left(\frac{r_{sc}}{l_c} \right)^4 G_{\max} > \begin{cases} (r_{sc} q)^4 F_{\max}^{(c)} & \text{charge fluctuations,} \\ 16(\varepsilon_F/\varepsilon_G)^2 F_{\max}^{(e)} & \text{energy fluctuations,} \end{cases} \quad (13)$$

in which $G_{\max} \approx 30$ (see Fig. 1), $F_{\max}^{(c)} \approx 0.2$, $F_{\max}^{(e)} \approx 0.98$; see Ref. 12. Since the maximum values $F_{\max}^{(c,e)}$ entering into the right side of (13) are attained at $\omega/qv_F \approx 0.68$ (for charge fluctuations) and 0.49 (for energy fluctuations), while the left side of (13) reaches its maximum at $(\omega/v_F)l_c \approx 1.93$, the inequality $ql_c \ll 1$ implies that the mechanism discussed here gives rise to a peak in the light scattering with a considerably larger change in frequency (this feature of the scattering spectrum should be observable when inequality (13) is violated as well).

For $\text{Al}_{0.2}\text{Ga}_{0.8}\text{As}$ with nonuniformity parameters $\bar{\delta} \approx 0.02$ and $l_c \approx 10^{-6} \text{ cm}^{-1}$ (Ref. 8), the mechanism discussed here turns out to dominate for concentrations of order 10^{17} cm^{-3} at excitation lengths close to $1 \mu\text{m}$. In this case, the ranges of concentrations n and correlation lengths l_c where inequality (13) is satisfied are found to be rather narrow [an additional limitation arises because of the condition that the nonuniformities at the bottom of the conduction band be totally screened, i.e., $(r_{sc}/l_c)^2 \ll 1$] and it is more convenient to study the scattering of far IR light (since high-power excitation by a CO_2 laser can be used). In the IR region, scattering by effective-mass nonuniformities predominates over a wide range of concentrations $n = 10^{16}$ to $3 \cdot 10^{17} \text{ cm}^{-3}$ and $l_c = 6 \cdot 10^{-7}$ to $5 \cdot 10^{-6} \text{ cm}$.

Experimental studies of Raman scattering spectra in nonuniform semiconductor alloys focus attention on the relation between the nonuniformity parameters and the shift and broadening of the LO phonon scattering line,¹³ whereas scattering by free carriers has been analyzed only in isolated papers (see, e.g., Ref. 14); the dependence of the spectra on the character of the technology-induced disorder is not discussed in these papers. The study of light scattering by unscreened electron effective-mass nonuniformities can be of interest for determining the compositional nonuniformity parameters of semiconductor alloys when l_c is small, where ordinary methods of investigation (x-ray, etc.) are inapplicable.

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¹⁾The fine-scale nonuniformities of the potential energy (which are caused by the statistical nature of the alloy and vary over distances of order of the lattice constant) are also unimportant in describing slow electrons near the conduction band extremum in the virtual-crystal approximation.⁵

²⁾If the contribution of the mechanism mentioned here is not dominant, but rather is comparable to contributions from other mechanisms, then in calculating the charge induced by the potential $\Phi(k)$ it is necessary to include the compositional nonuniformity. In this case, interference contributions arising from the fluctuation correction to $\varepsilon(\omega, \mathbf{k})$ are of the same order as the terms included in (7) and (8), which can change the spectral dependence of the single-particle scattering (9).

³⁾Single-particle scattering by spin density fluctuations gives a contribution of the same order of magnitude (but is observed only when the polarizations of the incident and scattered waves are crossed; see Refs. 1,2); therefore, the conditions for neglecting this mechanism are anal-

ogous to those discussed in the text.

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