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Atomic Radial Distribution Functions of As-Se Glasses

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X-ray-diffraction data from a series of arsenic-selenium glasses with compositions in the range Se-50-at.% As have been used to obtain atomic radial distribution functions for these materials. The first-neighbor correlation peak shifts linearly from 2.34 A for Se to 2.41 A for 36-at.% As and remains constant at higher As concentrations. Peak areas increase linearly with arsenic concentration and the magnitude agrees well with a model which assumes twofold coordination of selenium and threefold coordination of arsenic. At 40-at.% As and at 50-at.% As the first-neighbor peak is very similar to that obtained from the crystals As_2Se_3 and As_4Se_4 . However, beginning with the second-neighbor peak at about 3.70 \AA , there are significant differences between the crystalline and amorphous distributions. There is a smaller second peak in the glasses, the third crystalline peak is completely absent, and there are shifts in the positions of higher-order shells. Consequently, structural models based on microcrystalline regions of As_2Se_3 for 40-at.% As or As_4Se_4 for 50-at.% As did not give satisfactory agreement with the experimental distributions. In particular, the results do not support the existence of As_2Se_3 layers in the 40-at.% As structure.

INTRODUCTION

Previous studies of the structures of ehalcogenide glasses in the arsenic-selenium system have, with the exception of pure selenium, been restricted to suggestions of general models which might account for observed physical properties. In studies of the viscosity of a series of As-Se glasses Nemilov and Petrovskii¹ found an activation entropy dependence on composition which indicated three distinct regions, suggesting that at low arsenic concentrations the structure was primarily that of amorphous selenium, while at concentrations above 20-at. % As an As_2Se_3 type structure predominated, with a mixture of the two structures in the region $10-20$ -at. $%$ As. A similar result was obtained by Myers and $Felly²$ in a study of glass transition temperature T_g vs composition. Three distinct regions were indicated, with T_{ϵ} increasing with composition to 43at. $%$ As. This was attributed to the addition of arsenic to form branched selenium chains with, however, no significant As_2Se_3 -type network formation until 10-at. % As was reached. From 10 to 30-at. $%$ As, network formation begins and the two structures coexist, while above 30-at. $%$ As an essentially continuous network is formed with increasing continuity up to the stoichiometric composition. These results were also supported by Baidakov³ in an investigation of the paramagnetic contribution χ_p to the magnetic susceptibility of these glasses. An initial decrease in χ_p was associated with arsenic incorporation at selenium chain ends. At 10-at. % As a sharp rise in χ_p was attributed to the formation of a new type of structural unit, AsSe_{3/2}, which subsequently associated until, at 20-at. $%$ As and above, they formed an As_2Se_3 -type network.

Somewhat more direct evidence of the existence of specific types of arrangements is afforded by the infrared and Raman spectroscopic data of Schottmiller $et al.$ ⁴ Their work indicates that as arsenic is added there is a linear decrease in the concentration of Se_8 ring structures and a simultaneous growth in characteristic amorphous As_2Se_3 spectra. This process continues until a composition of 20-at. $%$ As is reached, at which point all Se_s ring structures have disappeared.

More definitive information, particularly for the 40-at. % As composition, has been gathered from studies of elastic moduli,⁵ heat capacity, 6 rrom studies of elastic moduli, heat capacity,
and infrared and Raman spectroscopy.⁷ In general, these studies reveal a great similarity between the amorphous and crystalline environments and are interpreted as implying a network structure. However, Kolomeits⁸ has cited the relatively low viscosity of the 40-at. % As composition as evidence of a predominantly chain or layer structure, rather than a network formation. .

Except for Se, only two previous radial distribution analyses of these glasses have been reported. Vaipolin and Porai-Koshits^{9–11} studie 40-at. $%$ As-60-at. $%$ Se, and on the basis of an increase in first-neighbor coordination, described a structure based on As_2Se_3 layers as found in crystalline As_2Se_3 but joined at points where As is in a nearly octahedral environment. However, their data were limited and the first coordination shell was not entirely resolved, so that these results must be considered tentative. Krebs and Schultze-Gebhard¹² investigated glassy 50-at. % As and found good agreement with a combination of radial distribution functions for amorphous Se and amorphous As. However, they could not determine whether the material was actually separated into pure Se and As regions, or whether a molecular structure such as As_4Se_4 was present. Se has been the subject of many studies¹³ and will not be considered in great detail here.

EXPERIMENTAL METHODS

A. Apparatus and Materials

Samples of As-Se glasses in the form of coarse powders were obtained from the Xerox Research Laboratory. They mere prepared from elemental As (99.999%}and Se (99.9999%}, manufactured by Gallard Schlessinger. The samples were placed in the proper proportion in quartz ampoules, which were evacuated to 10^{-6} torr, sealed, and melted at approximately 600'C for periods from 8 to 24 h. The furnace was equipped with a rocking mechanism to ensure thorough mixing of the elements. The ampoules were then allowed to cool in air (the pure selenium sample was quenched in ice water).

The samples were ground to 200 mesh and pressed at about 2000 lb/in.² to form briquettes $1 \times \frac{1}{2} \times \frac{1}{8}$ in. When not in use, samples were kept cool and dark to minimize crystallization and oxidation. Immediately prior to the diffraction studies, the samples were checked for crystalline phases, including oxides, the surface material was removed, and the briquettes were reground, pressed, and rechecked. While some oxide was visible before treating the surface, none was observed afterwards. Subsequent checks of composition agreed with the stated compositions to at least l at. % and indicated no detectable oxide. The densities of unpressed powders were determined by displacement employing paraxylene. The values obtained, shown in Fig. 1, agree well with those of Chernov, Dembovskii, and Chistov⁵ and were used in the subsequent analysis.

Two radiations were used in order to cover a maximum reciprocal-space range. For the region $0.4 \leq k \leq 6.0 \text{ Å}^{-1}$ ($k = 4\pi \sin \theta / \lambda$), CoK α radiation with a LiF monochromator in the incident beam was used. A proportional counter and singlechannel analyzer eliminated $\lambda/2$ components. For the region 2. $0 \le k \le 19.0$ Å⁻¹, RhK α radiation with a highly oriented graphite monochromator in the diffracted beam was used. A scintillation counter and pulse-height analyzer were employed, but $\lambda/2$ components were eliminated by keeping the operating voltage below 41 kV.

For amorphous samples the widest possible slit system was used to maximize intensities consistent with reasonable resolution. This resulted in about 0.5 $^{\circ}$ horizontal divergence. Soller slits were employed to give 3 $^{\circ}$ vertical divergence Low-index Ag-powder peaks had a full width at half-maximum (FWHM) of 19' with Rh and 36' with Co. All peak positions were accurate to 3' of calculated values.

Data were taken by point counting, using an approximate interval of 0.05 in k for amorphous samples. At least two passes were made over all angular ranges in opposite directions. Total counts

 (1)

exceeded $10⁴$ per point for all amorphous samples. The point-count data was recorded on paper tape and was later transferred to computer cards.

In addition to the amorphous samples, diffraction patterns from polycrystalline samples of As_2Se_3 and As_4Se_4 were also obtained. For these materials higher resolution in the region covered .by the cobalt tube mas required. Accordingly, slit widths were reduced to give 15' FWHM and the step size was reduced to 0. 01. In this case a minimum of 10^3 counts per point were accumulated in background regions. The analysis for these materials is the same as that for amorphous samples.

B. Analysis of Data

The distribution of atoms about an average atom in an amorphous material is related to the coherent scattered intensity by the usual Fourier integral

 $G(r) = 4\pi r [\rho(r) - \rho_0] = (2/\pi) \int_0^{\infty} F(k) \sin kr \, dk$,

with

$$
F(k) \equiv k(I - \langle f^2 \rangle) / \langle f \rangle^2 \tag{2}
$$

and

$$
\rho(r) \equiv \sum_{i} x_i K_i K_j \rho_{ij}(r) \tag{3}
$$

In these relations $G(r)$ is referred to as the reduced distribution, $F(k)$ as the reduced intensity, and we let $k=4\pi \sin\theta/\lambda$, $\langle f^2 \rangle$ is the $\sum_i x_i (f_i f_i^*)$, $\langle f \rangle^2$ $=(\sum_i x_i f_i)(\sum_i x_i f_i^*)$, $\rho_{ij}(r)$ = number of atoms/ \AA^3 of type j at a distance r from an average atom of type i, x_i is the atomic fraction of species i, ρ_0 is the average atomic density, f_i is the complex scat-

FIG. 2. Corrected reduced intensity $F(k)$ for some As-Se glasses.

tering amplitude of species i, and $K_i \equiv f_i / \langle f \rangle$.

For the As-Se system the assumption that K_tK_t is independent of k, necessary for deriving Eq. (1) , is well satisfied. Values of f_i , were taken from tables of Doyle and Turner 14 and Cromer.¹⁵

 $I(k)$ was obtained from the measured intensity, after background, polarization, and multiple scattering corrections, by normalizing at high k to $\langle f^2 \rangle + I_c$, where I_c is the incoherent scattering, Since a post specimen monochromator was employed for $RhK\alpha$ radiation, it was necessary to determine an incoherent scattering acceptance factor. This involved a mid- k normalization followed by subsequent corrections for slowly varying errors as described by Kaplow et $al.^{16}$. The acceptance factors were determined for each sample and successive values agreed to within 5% . Since the over-all correction for incoherent scattering did not exceed 25% of total intensity, the correction is good to 1%. In addition, the normalization constant mas verified following the integral method of Norman 17 and agreed with the former value to $0.5%$. The dependence of the incoherent scattering on k indicated an excess scattering at high k . That is, instead of falling smoothly to zero, it remained constant and equa to about $\frac{1}{2}$ the theoretical Compton incoherent scattering value above $k \approx 10 \text{ Å}^{-1}$. A similar excess incoherent scattering was reported by Warren and Mozzi, 18 although the magnitude was not indicated. It is possible that this arises from the continuous background, especially in view of the high tube voltage and rather broad mosaic of the graphite monochromator.

The reduced intensity $F(k)$ was extended to eliminate termination effects and corrected for slomly varying errors, following the methods of Kaplow et al , ¹⁶ These corrections resulted in changes of about 10% in first-peak heights, and less than 2% in first-peak areas.

DISCUSSION

Figures 2 and 3 show the reduced intensities $F(k)$ and reduced distribution $rG(r)$ for the materials studied. Only four of the intensity functions have been included, as the variations with composition are rather subtle. [Values for $F(k)$ and $rG(r)$ are available from the authors on request.] The areas and positions of the first and second peaks of the radial distribution function $4\pi r^2 \rho(r)$ are given in Table I.

The first peak, in the region from 2 to 3 \AA , increases linearly in areas with arsenic concentration. The magnitude of the area is consistent at all compositions with a model which assumes As to be threefold coordinated and Se to be twofold coordinated. For most compositions the area is about 2% greater than would be obtained from

TABLE I. Coordination numbers and distances for

Composition	$r_1(\AA)^a$	A_1^b	$r_2(\AA)^c$	A_2^d	2A ₂
Amorphous selenium	2,347	2.07	3.71	3.48	6.96
Monoclinic selenium	2.34	2.0	3.72	.	\cdots
$3-at.$ % As $-97-at.$ % Se	2.354	2.10	3.74	3.86	7.72
6-at. $%$ As-94-at. $%$ Se	2.350	2.14	3.70	3.70	7.40
12-at. % As-88-at. % Se	2.366	2.20	3.68	3.56	7.12
18-at.% As-82-at.% Se	2.374	2.26	3.69	3.77	7.54
21-at. % As-79-at. % Se	2.377	2.24	3.69	3.83	7.66
24-at. % As-76-at. % Se	2.382	2.27	3.68	3.91	7.82
30-at. % As-70-at. % Se	2.397	2.34	3.66	3.85	7.70
36-at.% As-64-at.% Se	2.412	2.38	3.65	4.08	8.16
40-at.% As-60-at.% Se	2.408	2.44	3.68	4.52	9.04
As_2Se_3	2.40	2.4	3.68	\cdots	9.20
45-at. % As-55-at. % Se	2.412	2.49	3.70	4.66	9.32
50-at. % As-50-at. % Se	2,408	2.55	3.73	4.82	9.70
As ₄ Se ₄	2.40	2.5	3.68	\cdots	9.50

 ${}^2\!r_1$ was measured to the midpoint of the half-maximum cord of the first peak in $4\pi r^2 \rho(r)$.

 ${}^{\text{b}}\!A_1$ was measured from the point at which the function $4\pi r^2 \rho(r)$ first became positive to the point where it next became zero.

 ${}^c\!r_2$ was measured to the maximum of the second peak in $4\pi r^2 \rho(r)$.

 A_2 was measured from the point at which the function $4\pi r^2 \rho(r)$ became positive for the second time to r_2 . It is one-half the symmetrized second-peak area.

such a model, but this is within the error of measurement and does not necessarily imply a small amount of excess coordination. For Se the deviation is about 3.5%, which is somewhat greater than experimentally allowable and may therefore be real. Several other authors have reported a similar value for the first-neighbor coordination of glassy selenium.^{12,19}

The peak position also shifts in a linear manner, at least between 6- and 36-at, $%$ As. There is a small increase in the very low As region, but the change is too slight to be considered significant. Such a linear change is in accord with Vegard's law for a solution of Se with 40-at. $%$ As-60-at. $%$ Se. However, this agreement would arise if phase separation were occurring as well. since the end points differ by only 3% and no peak separation can be hoped for as in a crystal. Thus this does not give any information regarding the formation of homogeneous solutions. However, beyond 36 -at. $%$ As there is no detectable shift in peak position. For 40-at, $%$ As this position, 2.41 Å, agrees with that for crystalline $As_2Se_3^{20}$ and our study of crystalline As.Se. shows that the position of the first-neighbor correlation for this material is also about 2.41 Å.

The second-neighbor peak shows a similar increase in area with composition. This increase is not linear, however, but close to parabolic. However, this area was only determined by considering the well-defined area on the low- r side. It would be possible to symmetrize the peak using

the low- r data, but this would just yield twice the area. There seems to be no reason to believe that such a symmetrical distribution might actually exist in the material, as the distribution in the crystal over the region of the second peak is not symmetrical. An increase in area is to be expected if arsenics acquire three nearest neighbors since such a configuration leads to an extra second neighbor, assuming the near neighbors of As to be Se. However, quantitative results for the second peak cannot be expected since it is impossible to meaningfully define its area or to separate atoms linked by shared near neighbors from those which are not so bonded. The position of the second peak shifts only slightly with a shallow minimum at around 30-at. % As. These results are also summarized in Table I.

Beyond the second-neighbor peak it is not possible to make quantitative assignments. However, there are regular variations in several features which would indicate their significance. The region from 4 to 5 Å shows a steady decrease in coordination, while from 5 to 6 Å there is a steady increase until 40-at. % As is reached and then coordination begins to fall. In the region from 6 to 7 Å the coordination approaches that of the average material till 21-at, $%$ As and then remains constant. The peak in the region from 7 to 8 \AA shifts toward smaller r , but does not change in size until about 40-at, $%$ As, when it begins to rise sharply. This could mean a relative depopulation of the region from 7.5 to 8 \AA . Such depopulation also occurs between 8 and 9 Å over the entire composition range. A peak in the $9-10-\text{\AA}$ region increases steadily to 40-at. $%$ As, but then falls. Beyond 10 \AA , peaks at 10.5 and 13.0 \AA of Se gradually change to minima, while a peak appears at about 12.5 Å .

There are also numerous small features which do not exhibit such regularity. It is felt that these are due to errors in $F(k)$, owing to the periodicity which many of them exhibit. In this regard the small peak in the region of 4.5 \AA in selenium might be due to such errors. However, it is considerably larger than other such features and a peak in this region has been reported by several authors, 13,21 It should be noted that the distribution function for selenium obtained in this study is in substantial agreement with that obtained by Kaplow. Rowe, and Averbach.¹³

MICROCRYSTALLINE MODELS

At the 40- and 50-at. % As compositions, a comparison with crystalline As₂Se₃ and As₄Se₄, respectively (see Fig. 4), shows that the first-peak areas and positions are identical and that the second-peak positions are the same. However, for both compositions, the amorphous second-peak area is less than that in the crystal and the third crystalline

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FIG. 4. Comparison of amorphous and crystalline atomic distributions. Upper, 50-at. % As-50-at. % Se and As₄Se₄; lower, 40-at. % As-60-at. % Se and As₂Se₃.

peak is absent altogether. That this peak is not simply broadened to the point that it is no longer distinguishable is indicated by the fact that the ratio of total crystalline area to total amorphous area continues to grow until about 5.0 Å , well beyond this peak. Thus it would appear that there is a depopulation of the region from about 4.0 to 5.0 \AA in passing from the crystal to its corresponding amorphous form. Beyond 5.0 \AA , amorphous correlation peaks appear and are located at somewhat smaller distances than similar crystalline peaks, in the region 5-10 A. Consequently the total area ratio decreases and begins to oscillate about the ratio of crystalline to amorphous density. Beyond 10 A it is difficult to determine a correspondence between crystalline and amorphous correlation distances.

An attempt was made to simulate the amorphous distributions at 40- and 50-at. $%$ As using microcrystalline models based on As_2Se_3 and As_4Se_4 . Experimentally determined distribution functions for the crystalline materials were damped by the function

$$
D(r) = 1, 0 - 1, 5r/d + 0, 5(r/d)^3
$$
 (4)

to account for the lack of correlation outside a sphere of diameter $d.$ ^{22,23} This procedure was used by Kaplow et al. 13 for amorphous Se. The diameter was chosen to give satisfactory damping at

15 A and to yield an approximately correct firstpeak height. For 40-at. $% A = 15$ Å; for 50-at. $% A = 15$ As, $d = 20 \text{ Å}$. It was found that even with this damping, corresponding peaks were either too sharp or too broad. Consequently the models were reconstructed using a distribution function calculated from the relation

$$
4\pi r \rho(r) = \sum_{i} \frac{C_i}{r_i (2\pi \sigma_{i})_{av}} \exp\left(-\frac{(r-r_i)^2}{2\sigma_i^2}\right), \qquad (5)
$$

where C_i is the coordination number at distance r_i , and σ_i^2 is the thermal broadening factor. The C_i are calculated for the known structure.²⁰ The σ_t^2 are related to the rms thermal displacement $\langle u^2 \rangle^{1/2}$:

$$
\sigma_i^2 \equiv \gamma_i \sigma_{\infty}^2 = \gamma_i \langle 2u^2 \rangle
$$
.

The γ_i are called coupling coefficients and express the degree to which the thermal vibration of atoms separated by a distance r_i are coupled. If $\gamma_i = 0$, then the thermal vibrations are coupled and there is no broadening of a correlation shell, the distance between the atoms remaining constant. If $\gamma_i = 1$, then $\sigma_i^2 = \sigma_{\infty}^2 = \langle 2u^2 \rangle$, the relative thermal vibrations are random, and there is a maximum broadening of the shell. This procedure has been developed by Kaplow and others in several papers. 13,16,24 Values for γ_i were determined by comparing our measured polycrystalline $G(r)$ for As_2Se_3 with the known structure.²⁰ For 50-at. $%$ As, values were determined in the processes of obtaining a structure for As_4Se_4 from our polycrystalline data. In both cases the value of σ_{∞}^2 determined was 0.061 \AA^2 . With this revised procedure for constructing the microcrystalline distribution, it is possible to vary the coupling coefficients to achieve approximately correct peak heights. In general this required that some γ_t be increased to values greater than 1.0, and this could imply that σ_{∞}^{2} was greater for the microcrystal than the extended crystal. The best distributions obtained in this way are shown in Fig. 5. It is obvious that the match with the amorphous pattern is not very good in either case. The primary problem is that peaks do not occur at the proper distances. Moreover, despite severe broadening, the third crystalline peaks are still visible. Additional problems occur at distances greater than 10 A, where the fit of the microcrystalline models is very poor.

The loss of the third-neighbor correlation peak is particularly significant in determining the kinds of changes which must occur in the crystalline structure to achieve the amorphous arrangement, For As_2Se_3 this peak occurs at 4.3 Å and is precisely the repetition distance along the c axis. Thus it corresponds to correlations almost entirely within the As_2Se_3 layer planes which lie perpendicular to the b axis. These layer planes may

FIG. 5. Comparison of microcrystalline models with experimental distributions.

be visualized as being constructed from either 12 member puckered rings or from As-Se spiral chains running along the c direction and c ross linked by Se. It is the repeat distance along this chain which is missing from the amorphous structure. Consequently it would appear that the As_2Se_3 layer structure must be distorted by stretching these spiral chains and thus distorting the layers. On the other hand, the existence of an interference maximum at $k = 1.25 \text{ Å}^{-1}$, which corresponds to the first crystalline interlayer interference for $As₂Se₃$, suggests that some type of regular structure exists at a separation of about 5 A. One possibility that meets both these criteria is the presence of As_2Se_3 chains. However, it is not possible to draw any certain conclusion about the existence of such a configuration from the analysis of radial distribution functions alone, and so network formation cannot be excluded. We are presently continuing work on the determination of atomic configurations for glasses in this system using Monte Carlo techniques, starting from crystalline and from random arrays.

For As_4Se_4 the third peak occurs at 4.5 Å. This distance is characteristic of correlations between atoms of neighboring As4Se4 molecules which comprise the structure. It may therefore be inferred

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that changes occur primarily by these molecules moving further apart, probably with additional distortions of the molecules themselves and with a randomization of their relative orientations.

CONCLUSIONS

The radial distributions determined for glasses of the As-Se system indicated a tendency for selenium to retain two nearest neighbors and arsenic three. In addition there is a gradual shift in the position of the average first-neighbor shell, which also agrees with the average of the sum of the covalent radii. This first shell appears to be clearly defined, at least for these airquenched bulk samples. Beyond the first shell there appear to be significant correlations out to about 12 Å for all samples. These results thus tend to support the conclusions of various autend to support the conclusions of various au-
thors^{1–4} regarding the effect of arsenic on selenium glasses, but imply that changes associated with threefold arsenic coordination occur in a much more gradual manner than would be indicated by some of these studies.

A comparison of the 40- and 50-at. % As correlations with distributions for the crystals As_2Se_3 and As_4Se_4 , respectively, indicates great similarities in near- neighbor coordination. This suggests that the local crystalline configurations, such as $Asso_3^{25}$ or $Asso_4$ molecules, also exist in the bulk glasses. However, differences beyond the first peak lead us to conclude that these materials cannot be considered to be microcrystalline without supposing such severe distortion of the crystalline lattices as to make the concept of doubtful utility. In particular, the existence of atomic layers such as those suggested by Tsuchihashi and Kawamoto²⁶ for amorphous As_2S_3 does not seem possible in bulk As-Se glasses.

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Self-Action of Laser Beams in Semiconductors*

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We have derived an expression for the nonlinear dielectric constant due to nonparabolicity of the conduction band and energy-dependent scattering processes in semiconductors under the influence of an external laser beam. The self-action phenomenon due to the combined effect is then studied. The role of linear and nonlinear absorption coefficients is incorporated in the analysis of self-action to show that (i), the former leads to a lower threshold value of the incident power of the beam for self-focusing to occur, and (ii) the latter decreases the self-focusing length, the reduction being appreciable at optical frequencies. Some properties of the self-action phenomenon are then described in practical situations. These considerations are applied in detail to a particular sample of n -InSb, where the absorption threshold is found to be significantly greater than the diffraction threshold and therefore, defocusing is favored. Finally, the possiblity of self-focusing, taking absorption effects into account, is discussed in elemental and group-III-V compound semiconductors-Ge, Si, GaAs, GaSb, GaP, InAs, InSb, InP, and AlSb — for visible He-Ne and infrared Q -switched CO₂ lasers.

I. INTRODUCTION

This paper is concerned with the theory of selfaction phenomenon (e.g. , focusing, defocusing, trapping, etc.) of laser beams in semiconductors and generalizes the earlier work of Tzoar and Gersten¹ and the present authors² to study the role of absorption processes in this phenomenon. It is expected that in semiconductors with the usual electron-hole concentration, the free-carrier effects would be much more important than other effects which give rise to self-action, such as the high-frequency Kerr effect, ³ electrostriction, ⁴ nonlinear electronic polarization, 5 self-induce thermal effects, 6 etc. In this paper, therefore, we have considered the two relevant mechanisms due to nonparabolicity (NP) of the conduction band¹ and the energy-dependent scattering² (EDS). While the NP mechanism is the dominant mechanism in low-band-gap semiconductors (e.g., InSb), the EDS mechanism is dominant in elemental semiconductors (e. g. , Ge and Si). However, irrespective of one mechanism being dominant over the other, the EDS mechanism besides giving rise to a nonlinear dielectric constant also accounts for the absorption of the beam inside the sample which has so far been neglected. Thus a complete account of the self-action phenomenon in semiconductors should be made by invoking both these mechanisms together, which is the aim of the present work.

In Sec. II we derive, using an elementary hydrodynamic approach, an expression for the nonlinear dielectric constant taking both the above-described