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Two-component electron-hole liquid: A simple model

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With the use of a very simple model for a uniaxially stressed semiconductor such as Ge or Si it is shown analytically under which conditions a phase separation of the electron-hole liquid into two distinct liquids of the type predicted by Kirczenow and Singwi would take place.

I. INTRODUCTION

Recent theoretical work[1-3] on the electron-hole liquid (EHL) in ⟨111⟩-stressed Ge by Kirczenow and Singwi predicted that in this system under suitable stress conditions the EHL at $T=0$ should phase separate into two liquids. One of the predicted phases consists of "cold" electrons and holes only, while the other phase contains "hot" electrons as well as "cold" electrons and holes. This prediction has apparently now found support in the experiments of Bajaj *et al.*[4] and of Timusk and Zarate.[5] In another publication[6] Kirczenow and Singwi conclude that in ⟨001⟩-stressed Si such a phase separation is much less likely to occur.

More recently Kirczenow[7] has made similar calculations of the properties of the EHL in ⟨110⟩-stressed Ge and Si. He predicts that in ⟨110⟩-stressed Ge the EHL should not phase separate at all, while in ⟨110⟩-stressed Si there should be no phase separation under experimentally accessible conditions. The above prediction is in marked contrast with that in ⟨111⟩-stressed Ge. Kirczenow attributes this difference in the behavior of the EHL in ⟨110⟩- and in ⟨111⟩-stressed Ge primarily to the different structure of the conduction band in the two cases. Kirczenow and Singwi[1] have attributed the nonparabolicity of the stress-split valence band as the primary cause of phase separation in ⟨111⟩-stressed Ge.

The aforementioned calculations are numerically fairly involved and therefore tend to obscure the underlying physics. Besides, in these calculations, the assumption that the exchange-correlation energy of the system depends only on the total density and is independent of the concentration of hot electrons has been made. Nonetheless, in view of the fact that the phase separation in question is the result of a delicate balance between kinetic and exchange-correlation energies, it is not quite certain whether a slight dependence of the exchange-correlation energy on concentration may spoil the final result.

We believe that the condensation into two distinct liquids that has been observed in ⟨111⟩-stressed Ge is a general phenomenon which, of course, exists only if the semiconductor has a "good" band structure: Uniaxial stress is just a device to make Ge achieve the latter. In this paper we wish to determine what are the main features of this good band structure in order to predict in which semiconductor one should look for the phase separation at hand. In order to examine this question in an analytical fashion, we shall take recourse to a simple model semiconductor.

II. SIMPLE MODEL OF A SEMICONDUCTOR

In a semiconductor with only *one* valence band and *one* conduction band, an *e-h* system is defined by two extensive variables: the volume V and the number N of holes (or electrons). The energy ϵ per *e-h* pair depends on one intensive variable $n=N/V$ and the curve $\epsilon(n)$ has one minimum, which gives the *e-h* liquid density. In order to have two plasmas, one needs to give more structure to the problem. The next simplest case is to condsider a semiconductor with *one* valence band of mass m and *two* conduction bands of masses m_L and m_R. This physically can describe not only the normal multivalley degeneracy but also some more complex

band structure. The *e-h* system is then defined by three independent variables: the volume V, the total number of electrons N and, for example, the number N_R of electrons in the "right" band. Neutrality of the charge implies that the number of electrons in the "left" band $N_L = N - N_R$. The energy ϵ per *e-h* pair now depends on two intensive variables $n = N/V$ and $y = N_R/N$. We shall see that n and y are indeed the relevant thermodynamical variables. The surface $\epsilon(n,y)$ presents a valley which corresponds to the minimum of ϵ vs n at a given y. If this valley has a saddle point in the y direction, or more generally a negative second derivative, then one expects that for a certain value of N_R/N around the saddle point, the energy of N holes and $N_R + N_L$ electrons is decreased if the *e-h* system splits into two plasmas with different values of y; one of which is larger than the initial y and the other smaller. If this happens then such a band structure with two different electron masses should exhibit the kind of phase separation we are seeking. The question is therefore: Is it possible to find values of m, m_R, and m_L for this saddle point to exist?

III. MATHEMATICAL CONSIDERATIONS

We shall be working at $T=0$. The total energy is

$$E(V,N,N_R) = N\epsilon(n,y) \ . \tag{1}$$

The system adjusts its volume to minimize its energy,

$$0 = \frac{\partial E}{\partial V}\bigg|_{N,N_R} = -n^2 \frac{\partial \epsilon}{\partial n}\bigg|_y \ . \tag{2}$$

This is the usual relation of the *e-h* droplet phase separation. The plasma density is given by the minimum of $\epsilon(n,y)$ at constant y. We shall represent this minimum by $\tilde{\epsilon}(y)$.

Let us assume for a moment that $\tilde{\epsilon}(y)$ has negative curvature [i.e., $\tilde{\epsilon}''(y) < 0$] somewhere in the interval $y_1 < y < y_2$; y_1 and y_2 being the points of contact with the common tangent as shown in Fig. 1. For a given $y = N_R/N$ lying in the interval $y_1 < y < y_2$, we immediately see that

$$\tilde{\epsilon}(y) > \bar{\epsilon}(y) \equiv N_1 \tilde{\epsilon}(y_1) + N_2 \tilde{\epsilon}(y_2) \ , \tag{3}$$

where

$$N_1 = N\frac{y_2 - y}{y_2 - y_1} \ , \quad N_2 = N\frac{y - y_1}{y_2 - y_1} \ , \tag{4}$$

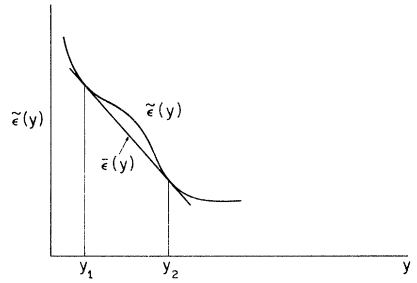

FIG. 1. Qualitative behavior of $\tilde{\epsilon}(y)$ vs y. y_1 and y_2 are the points of contact of the curve with the double tangent and determine the values of y in the two phases. $\bar{\epsilon}(y)$ is along the double tangent.

and

$$N = N_1 + N_2 \ .$$

This means that for this value y the system is unstable since it can decrease its energy by splitting into two plasmas with ratios y_1 and y_2 and containing N_1 and N_2 *e-h* pairs, respectively. It is easy to show that this double tangent construction ensures the equality of each of the two chemical potentials in the two phases.

The problem now is to find when the curve $\tilde{\epsilon}(y)$ of the minimum energy has a negative curvature to allow the double tangent construction. For that one has to find $\epsilon(n,y)$ for this model plasma. The kinetic part of the energy is

$$\epsilon_{\text{kin}}(n,y) = \frac{2.21}{r_s^2}\alpha(y) \ , \tag{5a}$$

where

$$\alpha(y) = 1 + \rho_R y^{5/3} + \rho_L (1-y)^{5/3} \ . \tag{5b}$$

In Eq. (5) the energy is in units of rydberg and the length is in units of Bohr radius, both units being associated with the holes (mass m) only. $\rho_{L,R}$ is the ratio $m/m_{L,R}$. Let m_L be the lighter of the two conduction masses. $\alpha(y)$ has a minimum for $y = y_c$ which corresponds to the equality of the Fermi energy of the two conduction bands—a result which is expected on physical grounds. $\alpha(y)$ as well as its first and second derivatives $\alpha'(y)$ and $\alpha''(y)$ are shown in Fig. 2. Note that $\alpha'' \to \infty$ when $y \to 0$ or 1.

The Coulomb interaction gives rise to exchange and correlation energies $\epsilon_{\text{Coul}}(n,y)$. We shall first follow Kirczenow and Singwi and assume that ϵ_{Coul} depends only on n. This assumption has been

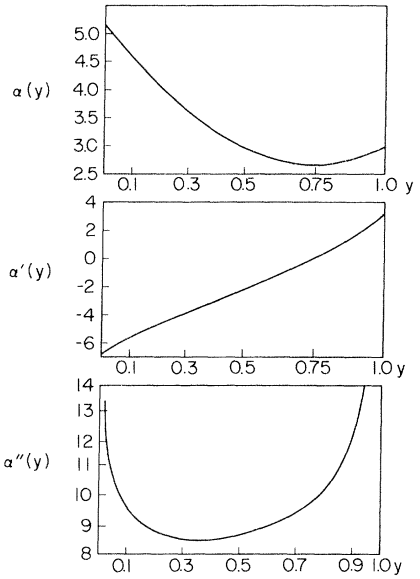

FIG. 2. $\alpha(y)$, $\alpha'(y)$, $\alpha''(y)$ vs y. The curves are calculated for the values of the masses (in electron mass unit): $m_L=0.22$; $m_R=0.458$; $m=0.916$. $\alpha(y)$ has a minimum at $y=y_c=0.75$.

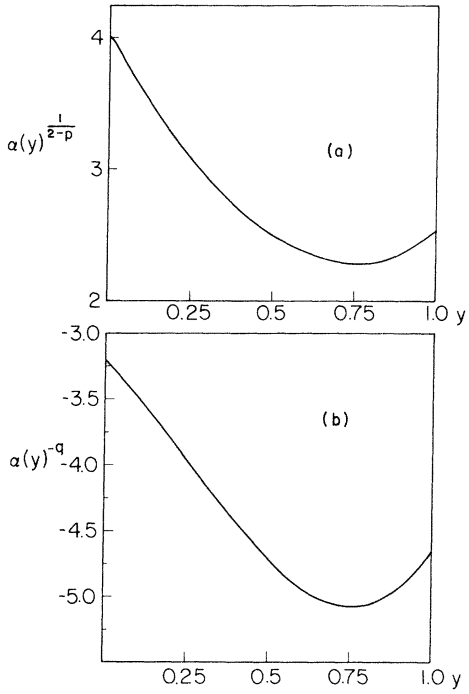

FIG. 3. (a) $\alpha(y)^{1/2-p}$ and (b) $\alpha(y)^{-q}$ vs y. The two quantities are, respectively, proportional to $r_{s0}(y)$ and $\tilde{\epsilon}_0(y)$. The masses are the same as those used in Fig. 2. p is taken to be 0.82. There is an interval of slightly negative curvature between $y=0.01$ and $y=0.25$ in Fig. 3(b).

empirically found to be very good. However, here one has to be a bit careful since one is interested in the curvature of $\tilde{\epsilon}(y)$, and a very good approximation on $\tilde{\epsilon}(y)$ may not turn out to be so good for $\tilde{\epsilon}''(y)$. We shall come back to this question in the next section. For the present, we shall take the Coulomb energy of the form

$$\epsilon^0_{\rm Coul}=-\frac{\beta_0}{r_s^p}\ , \tag{6}$$

where β_0 is a constant independent of y. The exponent p is equal to unity for exchange alone. In the density range of interest here, one finds[8] that for the exchange plus correlation energy p is in the range 0.7—0.8. The simple expression (6) has the advantage that it allows an analytic calculation which helps to investigate from where the instability arises. The total energy $\epsilon(n,y)$ per pair is then given by the sum of expressions (5) and (6).

The minimum of $\epsilon(n,y)$ at constant y is

$$\tilde{\epsilon}(y)=-\epsilon_0\alpha^{-q}(y)\ , \tag{7a}$$

corresponding to

$$\tilde{r}_{s0}(y)=r_0\alpha^{1/(2-p)}\ , \tag{7b}$$

where r_0 and ϵ_0 are constants independent of y, and $q=p/(2-p)$ is a number lying between 0 and 1. From $\alpha(y)$ we deduce that $\tilde{\epsilon}_0(y)$ and $\tilde{r}_{s0}(y)$ are minimum for $y=y_c$ (see Fig. 3). Around y_c, $\tilde{\epsilon}_0'(y)$ is positive. Phase separation into two e-h plasmas will exist if $\tilde{\epsilon}_0'(y)=0$ somewhere, i.e., if the following condition is satisfied:

$$\frac{\alpha'^2}{\alpha^2}=\frac{1}{q+1}\frac{\alpha''}{\alpha}\ . \tag{8}$$

Figure 4 shows the typical behavior of these two quantities as a function of y. In order to see what the masses should be for the solution to exist, one rewrites (8) in the form

$$A\equiv\frac{\alpha\alpha''}{\alpha'^2}=q+1\ , \tag{9a}$$

which on using (5b) and after some algebra becomes

$$A=\frac{2}{5}\left[1+\frac{f(\rho,\rho_R,t)}{t(1-t^2)^2}\right]=q+1\ , \tag{9b}$$

where

$$t=\rho^{-1/2}\left[\frac{1-y}{y}\right]^{1/3}\ ,$$

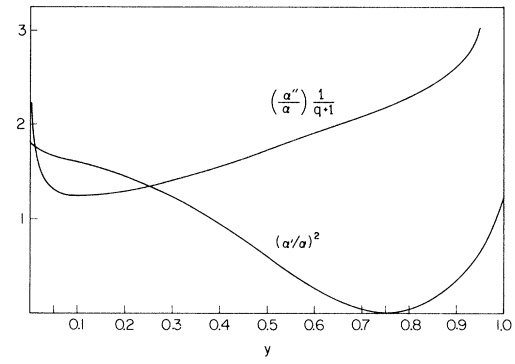

FIG. 4. Graphical solution of Eq. (8) for the same values of the parameters used in drawing the curves in Figs. 2 and 3.

$$f(\rho,\rho_R,t)=\rho^{-3/2}(1+\rho^{3/2}t^3)^2 \times\left[1+\frac{1}{\rho_R}\frac{1+\rho^{3/2}t}{(1+\rho^{3/2}t^3)^{1/3}}\right], \quad (10)$$

and

$$\rho=\frac{m_L}{m_R}<1 .$$

Now $\alpha\alpha''/\alpha'^2$ diverges for $y=0$, y_c, and 1, i.e., $t=\infty$, 1, and 0, respectively. A solution $\tilde{\epsilon}''(y)=0$ exists if $\alpha\alpha''/\alpha'^2$ is less than $q+1\sim 1.7$ somewhere. From Eq. (10) we see that $f(\rho,\rho_R,t)$ is an increasing function of t and is always larger than $\rho^{-3/2}(>1)$. We also note that $f(\rho,\rho_R,t)$ decreases with increasing ρ_R for constant ρ and t.

Case (i). $0<t<1$ (i.e., $1>y>y_c$)

In this range of t values, $1/t(1-t^2)^2$ is greater than 3.5 and since $f(\rho,\rho_R,t)$ is always greater than unity, the left-hand side of Eq. (9b) is always greater that 1.7, whatever be the values of ρ and ρ_R. Therefore a phase separation into two plasmas can never occur if the initial y is larger than y_c corresponding to the equality of Fermi energy in the conduction band. It can be proven very generally that this result is valid for values of $p\leq 1$ (see Appendix A).

Case (ii). $1<t$ (i.e., $y_c>y>0$)

In this case a phase separation can be shown to exist for suitable values of ρ and ρ_R. In Fig. 5 the relevant values of these two quantities are shown as shaded. The value of p is 0.82. One can say generally that large values of both m/m_R and m_R/m_L help the instability to occur.

The solid circle in Fig. 5 have been drawn for

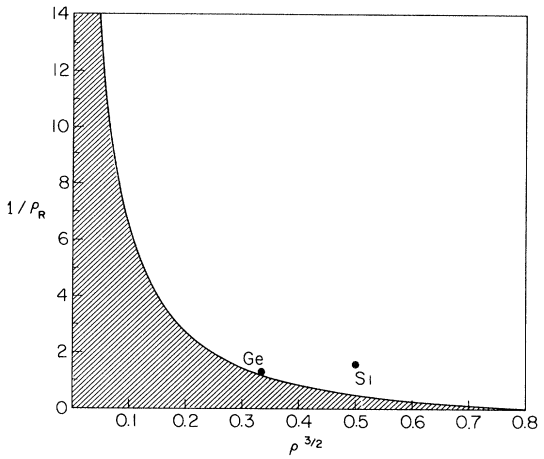

FIG. 5. Shaded region under the curve corresponds to the values of the quantities $\rho^{3/2}$ and ρ_R^{-1} for which a phase separation into two EHP exists: $\rho=m_L/m_R$, $\rho_R=m/m_R$. Points corresponding to Ge(111) ($m=0.347$, $m_L=0.22$, and $m_R=0.458$) and Si(100) ($m=0.523$, $m_L=0.510$, and $m_R=0.809$) as given by the simple model are shown in the figure. The value of p is 0.82.

Ge ($m=0.347$, $m_L=0.22$, and $m_R=0.458$) and Si ($m=0.523$, $m_L=0.510$, and $m_R=0.809$). It is seen that these points lie above the shaded region (Ge being on the border line); and hence this simple model fails to predict a phase separation even under the most favorable assumption for the hole masses. A more detailed discussion of this point is reserved for Sec. VI.

IV. EFFECT OF DEPENDENCE OF ϵ_{xc} ON y

In this section we shall examine the validity of the approximation (6). One knows from experience that the correlation energy has the effect of smoothing the dependence of the exchange energy on any parameter. However, qualitatively the combined behavior of exchange and correlation retains a "memory" of the exchange alone.[9] The latter is easy to calculate as a function of y and r_s. One finds that

$$\epsilon_x(y,r_s)=-\frac{\beta_x(y)}{r_s} , \quad (11a)$$

where

$$\beta_x(y)=0.916[1+y^{4/3}+(1-y)^{4/3}] . \quad (11b)$$

$\beta_x(y)$ has a minimum for $y=\frac{1}{2}$ and is a very flat

function of y, but its second derivative becomes infinite for $y=0$ and $y=1$ (as α''). We shall show in Appendix B that this divergent behavior of $\beta_x'(y)$ always leads to a phase separation in the Hartree-Fock approximation. As $\epsilon_{\rm Coul}$ countains ϵ_x, such a divergence will persist in $\epsilon_{\rm Coul}''(y)$, and this may affect the instability in question. To see the consequences of such a divergence, we write the *e-h* energy as

$$\epsilon(n,y)=2.21\frac{\alpha(y)}{r_s^2}-\frac{\beta(y)}{r_s^p}\ , \tag{12}$$

where $\beta(y)$ has the same qualitative behavior as $\beta_x(y)$ but only somewhat flatter. The behavior of β, β', and β'' is, respectively, like that of α, α', and α'' for $y_c\simeq\frac{1}{2}$. The minimum energy is then given by

$$\tilde{\epsilon}(y)=-\epsilon_1\beta\left[\frac{\beta}{\alpha}\right]^q\ , \tag{13a}$$

for

$$\tilde{r}_s(y)=r_1\left[\frac{\alpha}{\beta}\right]^{1/(2-p)}\ , \tag{13b}$$

where ϵ_1 and r_1 are independent of y. As $\beta(y)$ is a very flat function, $\tilde{\epsilon}$ and $\tilde{r}_s$ are *a priori* not expected to differ very much from $\tilde{\epsilon}_0$ and $\tilde{r}_{s0}$ given in Eq. (7). But if one looks at the possibility of a change in curvature of $\tilde{\epsilon}(y)$, one has to look for the solution of

$$\frac{\alpha'^2}{\alpha^2}+\frac{1}{q}\frac{\beta''}{\beta}+\frac{\beta'^2}{\beta^2}=\frac{1}{q+1}\frac{\alpha''}{\alpha}+2\frac{\beta'\alpha'}{\beta\alpha}\ . \tag{14}$$

Equation (14) replaces Eq. (8). One sees that the real structure of β may play a role. One can say that if for some values of the masses Eq. (8) has a solution, Eq. (14) will also have one, provided the new terms decrease the right-hand side of (8) and increase the left-hand side in the range of values of y for which the solution of (8) exists (see Fig. 4). One immediately sees that since β''/β and β'^2/β^2 are both positive, the curve α'^2/α^2 is pushed up by these new terms for all y. Since $\alpha'<0$ for $y<y_c$ and $\beta'<0$ for $y\le\frac{1}{2}$, $(1/q+1)(\alpha''/\alpha)$ is pushed down by the new terms for $\frac{1}{2}<y<y_c$. Therefore, if Eq. (8) had a solution for $\frac{1}{2}<y<y_c$, Eq. (14) will also have one. The dependence of β on y thus helps to realize $\tilde{\epsilon}''=0$. One, therefore, arrives at the conclusion that if a band structure gives an instability within the approximation of a Coulomb energy being independent of y, the existence of this instability will persist.

V. PHASE SEPARATION IN THE MODEL SEMICONDUCTOR

We shall now discuss how the preceding calculation applies to the possible observation of the coexistence of two EHP. We first start with the case of Ge under ⟨111⟩-uniaxial pressure as considered by Kirczenow and Singwi. Our "left" mass corresponds to the mass m_e of the conduction band, while our "right" mass is $m_R=3^{2/3}m_e$ (see Fig. 6). The latter takes care of the threefold degeneracy of the conduction band in stressed Ge. The shift Δ of the left conduction band does not play any role since the total number of electrons in it stays constant in the phase separation. In order to produce two EHP we have seen that one needs an initial y between y_1 and y_2 (Fig. 1). When the *e-h* are produced by the laser pulse at time $t=0$, by symmetry the upper three valleys and the lower one valley are filled to the same level, i.e., $y=y_c$, and the plasma does not split. As t increases, the electrons begin to fall into the lower valley, i.e., in the left band so that y decreases and eventually reaches the region $y_1<y<y_2$ where two EHP coexist. Note that the hole density of the plasma y_1 which contains more of the "light" electrons is smaller than the one of the plasma y_2 [i.e., $\tilde{r}_s(y_1)>\tilde{r}_s(y_2)$]. For Δ large enough, one explores the region $y\le y_c$ and sees the splitting of the initial EHP as t increases and as observed by Bajaj *et al.*[4]

It would be interesting if such a coexistence of two plasmas could be obtained as a quasi-equilibrium state in a CW experiment. This would imply an equilibrium y which is different from y_c. In all likelihood such a situation has been realized in the experiment of Timusk and Zarate[5] where they have observed a new EHD plasma resonance in stressed Ge corresponding to a second plasma containing both hot and cold electrons and which is of higher density.

For a multivalley structure without uniaxial stress all the electrons have the same Fermi momentum so that $y=y_c$ and only one plasma exists as in normal Ge and Si. y will be different from y_c only if the bottoms of the conduction bands are not at the same energy. This of course is realized when a uniform pressure is applied but also in alloys such as Ge-Si when the composition is such that the Ge and Si conduction bands are close.

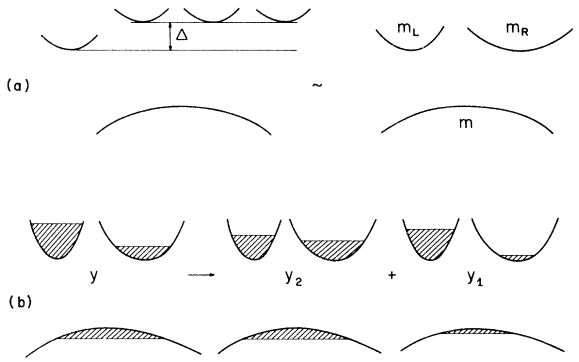

FIG. 6. (a) Band structure of a multivalley semiconductor under uniaxial stress is replaced by a model with only two inequivalent conduction bands. (b) Densities of the two EHP when phase separation occurs. This drawing is made within the free-particle model; the Coulomb interaction, of course, shifts the free energies so that the chemical potentials of each of the two kinds of electrons are the same in the two plasmas.

VI. CRITIQUE OF THE MODEL

The simple model of an indirect band semiconductor that we have discussed in the foregoing sections involves only two parameters, one the ratio $\rho_R = m/m_R$ and the other the ratio $\rho = m_R/m_L$; and given the values of these parameters it has the merit of predicting in a simple and analytic way the conditions under which two *e-h* plasmas would coexist. In a real situation such as ⟨111⟩-Ge or ⟨100⟩-Si, the model is capable of giving only approximate answers. The reason for this is that one does not have a reliable method of estimating the hole mass m of the single parabolic valence band which replaces the complex structure of two coupled hole bands in the stress region of interest. For example, in Ge-⟨111⟩ we know through detailed calculations[1] that a phase separation exists in the region of stress when the Fermi energy of holes is comparable to the stress splitting. However, in the present simple model for the phase separation to exist $1/\rho_R$, i.e., m_R/m has to be less than 1.21 (since $\rho^{3/2}=0.33$). This is not the case even if we take for the hole mass a value corresponding to that of the heavy hole in normal Ge (see Fig. 5). This condition is even more violated if we take the mass of the hole to be that corresponding to the infinite stress limit of Ge-⟨111⟩. In the case of ⟨100⟩-Si the model even in the most favorable circumstances does not predict a two-phase separation. There is no simple way of extrapolating the hole mass in the stress region of interest. Nonetheless, the model is capable of making some definite predictions. For example, if $\rho^{3/2}$ exceeds 0.8 (see Fig. 5) there can be no phase separation into two plasmas. This will be the case for ⟨110⟩-Ge. Detailed calculations[7] of Kirczenow do support this prediction. As long as $\rho^{3/2}<0.8$ and $1/\rho_R$ is very small, i.e., hole mass very large, the model always predicts the coexistence of two plasmas.

VII. CONCLUSIONS

In a semiconductor having large m/m_R and m_R/m_L ratios, an *e-h* plasma splits into two separate plasmas when the initial Fermi energy of the light conduction band is larger than that of the heavy one (Fig. 6). This splitting can be visualized as most of the heavier electrons with some light ones concentrating in a region having a large hole density, while the rest of the light electrons with their holes form a second plasma at lower density (in order to keep a small Fermi energy).

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APPENDIX A

Here we prove that a phase separation into two plasmas can never occur if $y \geq y_c$ for any value of p in the interval $0 \leq p \leq 1$. This is mathematically equivalent to proving that [see Eq. (9b) of the text]

$$A \geq q+1 \quad \text{for } 0 \leq t \leq 1 , \tag{A1}$$

with

$$0 \leq p \leq 1, \quad 0 < \rho_R < \infty ,$$

$$0 < \rho^{3/2} \equiv s < 1 .$$

This holds if

$$f(\rho,\rho_R,t) \geq 4t(1-t^2)^2 . \tag{A2}$$

From Eq. (10) we see that

$$f(\rho,\rho_R,t) > s^{-1}(1+st^3)^2$$

$$\equiv g(s,t) > g(1,t) = (1+t^3)^2 ,$$

where we have used the fact that $g(s,t)$ is a decreasing function of s for $0 \leq t \leq 1$. Therefore, inequality (A2) is satisfied if

$$(1+t^3)^2 \geq 4t(1-t^2)^2$$

or

$$(t^2-3t+1)^2 \geq 0 .$$

This completes the proof.

APPENDIX B

Here we show that the Hartree-Fock (HF) approximation always predicts a phase separation for $y \to 0$ and $y \to 1$ irrespective of the parameters of the band structure. In the HF approximation, the energy per pair is given by

$$\epsilon(n,y) = \frac{2.21}{r_s^2(n)}\alpha(y) - \frac{\beta_x(y)}{r_s(n)} ,$$

where

$$\alpha(y) = 1+\rho_R y^{5/3}+\rho_L(1-y)^{5/3} ,$$

$$\beta_x(y) = 0.916[1+y^{4/3}+(1-y)^{4/3}] .$$

Now

$$\tilde{\epsilon}(y) = -\epsilon_1 \beta_x^2(y)/\alpha(y)$$

and

$$\tilde{\epsilon}''(y) = -\epsilon_1 \left[\frac{\beta_x'^2+2\beta_x\beta_x''}{\alpha(y)} + 2(2\beta_x\beta_x') \left[\frac{\alpha'}{\alpha^2} \right] + \beta_x^2 \left[\frac{\alpha''}{\alpha^2} + \frac{2\alpha'^2}{\alpha^3} \right] \right] .$$

When $y \to 0$ or 1, the behavior of $\tilde{\epsilon}''(y)$ is determined only by the most divergent term, i.e.,

$$\tilde{\epsilon}''(y) \sim -2\epsilon_1 \frac{\beta_x(y)\beta_x''(y)}{\alpha(y)} < 0 .$$

Therefore, in the HF approximation there is always a phase separation.

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