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Publication Date

1975-09-01

Submitted to the Journal of Physics and Chemistry of Solids

LBL-4131 Preprint c

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Prepared for the U. S. Energy Research and Development Administration under Contract W-7405-ENG-48

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Proposed Experiment to Realize and Detect the Three Dimensional Wigner Lattice

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ABSTRACT

This paper suggests and discusses possible experimental means of realizing and detecting the three dimensional Wigner electron lattice. It is concluded that very pure germanium, doped with antimony, offers the possibility of obtaining extrinsic electron densities low enough to form the Wigner lattice. It is proposed that Brillouin scattering of photons by the LA and TA modes of vibration of the lattice offers a method of experimental detection. This is because the Brillouin scattering spectrum of the electron lattice is anticipated to include structure not found in the spectrum of the electron gas of the same density.

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I. Introduction

The Wigner lattice [1] is a body-centered cubic lattice of electrons whose formation in a solid is predicted to occur at a sufficiently low value of the electron density. With the present availability of very pure semiconductors with variable extrinsic electron densities, it is interesting to consider the possibility of experimentally realizing and detecting the three-dimensional [2] Wigner lattice. The aim of this paper is the suggestion and discussion of possible experiments for this purpose. To the best of the author's knowledge, this is the first paper discussing the possible experimental realization and detection of the three-dimensional Wigner lattice.

Considering an assembly of electrons in the presence of a uniform background of fixed positive charge, Wigner [1] argued that, as their potential energy became large compared to their kinetic energy, the electrons would become localized in space. This would make the kinetic energy ineffective in opposing this spatial localization and lead to the formation of a lattice of electrons. If n is the electron density in a solid of dielectric constant ε , then the potential energy V of interaction of the electrons is roughly

$$V \approx (-e)^2 / \varepsilon r_o$$

where r_o is the average distance between electrons. Making the Wigner-Seitz approximation, $(4\pi r_o^3/3) = (1/n)$, the potential energy may be

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expressed as

$$V = (4\pi/3)^{1/3} (e^2/\epsilon) n^{1/3}$$

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The average kinetic energy T of an electron may be taken as equal to (3/5) of the Fermi energy $E_{\rm F}^{}$, so

$$T \approx \frac{3}{5}E_F = (3\hbar^2/10m)(3\pi^2n)^{2/3}$$

where m is the free electron mass. Then the ratio

$$(V/T) = (4\pi/3)^{1/3} [10/3(3\pi^2)^{2/3}] (me^2/\hbar^2 \epsilon) n^{-1/3},$$

and (V/T) will become large at low values of the electron density n. Defining an effective Bohr radius [3] a_0^* by the relation

$$a_{o}^{*} \equiv (\hbar^{2} \epsilon / m^{*} e^{2}),$$

where we are now considering electrons of effective mass m^{*}, one obtains

$$(V/T) = C(r_0/a_0^*) \equiv Cr_s$$
(1)

where the constant C = $(10/9)(16/3\pi^2)^{1/3} = 0.905$. The expression (1) shows that (V/T) increases as the dimensionless parameter r_s increases,

leading to the crystallization of the electron gas at sufficiently large values of r_s , say larger than a critical value [4] of $r_s = r_c$.

The ground state energy of the electron gas and lattice has been the subject of a number of theoretical calculations [4-8]. The values of r_c obtained are generally between 14 and 20, leading to the expectation that the Wigner lattice will be stable at electron densities corresponding to values of r_s greater than about 15. Large values of r_s are favored by the low values of the electron density which can be achieved in extrinsic semiconductors at low temperatures. It is thus natural to consider such semiconductors as potential "hosts" in which to examine the possibility of achieving and detecting the Wigner electron lattice.

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II. Possible Formation of the Lattice

The semiconductor presently obtainable in the purest form is germanium, in which net carrier concentrations of 10^9 to 10^{11} cm $^{-3}$ may be obtained[9]. The existence of germanium of such high purity suggests that extrinsic electron concentrations low enough to produce the Wigner lattice may be achievable. Using an effective mass value[10] $m^* = 0.12m$ for electrons and a dielectric constant $\varepsilon = 16$, equation (1) may be used to calculate values of r_s as a function of the electron density n in germanium. If a typical theoretical value of $r_c = 15$ is chosen, it is found that r_s is larger than 15 if the electron density n is less than 8.7 x 10^{14} cm⁻³. Considering antimony as a donor in germanium (with an ionization energy $E_{A} = 0.0096 \text{ eV}[11]$), one may calculate [12] the electron concentration n as a function of temperature for a given value [13] of the donor atom concentration N_d. If N_d = 10^{16} atoms of antimony per cm³, the electron density n is less than 3.5 x 10^{14} ${\rm cm}^{-3}$ for temperatures below 16K. Such conditions are readily achievable in the very pure germanium available today. It thus appears that values of r larger than the calculated values of the critical value r may be obtained for the extrinsic electron gas in antimony doped germanium [14].

We next calculate some parameters of the electron gas at a low density. Using a representative value of the electron density $n = 10^{14}$ cm⁻³, corresponding to $r_s = 19$ in germanium, the lattice constant a of the bcc electron lattice is given by $n = (2/a^3)$, or $a = 2.7 \times 10^{-5}$ cm. It is well known [15] that the electron gas in a semiconductor of simple band structure [16] has a plasma oscillation mode of frequency ω_p , where $\omega_p^2 = (4\pi ne^2/m*\epsilon)$. For germanium with $n = 10^{14}$ electrons per cm³, m* = 0.12m, and $\epsilon = 16$, a value of $\omega_p = 4.1 \times 10^{11} \text{ sec}^{-1}$ is obtained, corresponding to a $\Gamma(k=0)$ plasmon energy $\hbar \omega = 0.269 \times 10^{-3} \text{ eV} = 0.27 \text{ meV}$.

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<u>III. Possible Light Scattering Experiment to Detect the Lattice</u> Assuming that the Wigner lattice does form under the conditions suggested, how might it be detected experimentally? It is well-known [17] that plasmons in the conduction electron gas in semiconductors have been detected by light (Brillouin) scattering experiments. In such experiments, an incident photon of frequency ω_0 is inelastically scattered with the creation of a plasmon of frequency ω_p and a scattered photon of frequency ω_s . The result is a frequency shift $\Delta \omega \equiv (\omega_0 - \omega_s) = \omega_p$ for pure plasmon scattering without plasmon-phonon interactions [17]. For example, such Brillouin scattering has been observed [18] in n-type GaAs. In this case, the observed plasma frequency $\omega_p \approx 100 \text{ cm}^{-1}$, corresponding to a plasmon energy $\hbar \omega_p = 1.2 \text{ meV}$, at an electron concentration of about 1.16 x 10^{17} cm^{-3} .

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If the conduction electron <u>gas</u> in a semiconductor does crystallize into an electron <u>lattice</u>, it would be expected that <u>transverse</u> acoustic [19] vibrations would be possible in addition to a longitudinal acoustic branch. This expectation is borne out in the calculation by Clark [20] of the vibrational spectrum of an electron lattice in the presence of a uniform background of positive charge [21]. Clark calculates the frequencies of both longitudinal (LA) and transverse (TA) acoustic modes as a function of wave vector for several directions of propagation in the bcc electron lattice. The LA mode is unusual in that its frequency is not zero at the zone center; this is due to the long range of the Coulomb potential. (This behavior of the vibrational spectrum is the same as that observed for plasma oscillations in the electron gas [22].) The electron lattice thus has vibrational modes whose phonon aspects may be thought of approximately as longitudinal and transverse "plasmons". The former are the analog of longitudinal plasma oscillations in the electron gas, but the latter should be something new.

Clark's result is that the frequencies ω_{ℓ} and ω_{t} of the LA and TA modes are given by

 $\omega_{\ell} = (1/\sqrt{2\pi})\Lambda_{\ell}\omega_{\mathbf{p}} \qquad ; \qquad \omega_{t} = (1/\sqrt{2\pi})\Lambda_{t}\omega_{\mathbf{p}},$

where $\omega_p^2 = (4\pi ne^2/m*\epsilon)$ is the plasma frequency of the electron gas of density n, etc. The quantities Λ_g and Λ_t are reduced frequencies whose calculated values [20] at various special points [23] of the Brillouin Zone are given in Table I. While the frequency of the zone center LA mode is the same as the plasma frequency of the electron gas of the same density, the zone edge (points H and P) LA and TA modes of the electron lattice are calculated to have frequencies of about 0.6 ω_p .

Table I. Vibrational Frequencies of the bcc Electron Lattice.

· ···	Special Point	Γ(000)	H(100)	P(111)	
•	• •	•		- · · ·	
· · · ·	۸ _e	2.50	1.45	1.45	
۰.	۸ _t	0.0	1.45	1.45	·
	$(\omega_{\ell}/\omega_{p})$	1.00	0.58	0.58	
	(ω_t/ω_p)	0.0	0.58	0.58	
	• -			•	

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One may contrast the expected features of the Brillouin scattering [24] spectrum of the electron lattice with those observed in Brillouin scattering by plasmons in the elctron gas. In the latter case, the central feature of the spectrum is [18] a peak at the photon frequency shift $\Delta \omega = \omega_{n}$ due to scattering by the plasmons which are the LA vibrational mode of the electron gas. For the electron crystal, one would also expect to observe Brillouin scattering at a photon frequency shift $\Delta \omega = \omega_{p}$ due to the zone center LA vibrational mode whose calculated frequency is ω_{p} . However, it is also to be expected that the additional vibrational modes possible due to the formation of the electron lattice would result in new and additional features in the Brillouin scattering spectrum, and that these features would not be present in the equivalent spectrum for the electron gas. Of special interest are the transverse vibrations of the electron lattice; these would be expected to couple strongly to incident photons. However, for these TA modes, the zone-center frequency is zero, so it is of particular interest and importance that, for the electron lattice, one would expect Brillouin scattering by LA and TA modes whose wave vectors are significantly different from zero.

The reason for this is the fact that the lattice constant a of the electron lattice is very large compared to the values of the lattice constant found in most crystals. This results in the electron lattice having a much smaller Brillouin Zone than those usually encountered. For an electron density $n = 10^{14} \text{ cm}^{-3}$, the lattice constant a is

(2)

approximately 2.7 x 10^{-5} cm, so the Brillouin Zone boundary (π/a) is at a wave vector about equal to 10^{5} cm⁻¹, a value about three orders of magnitude smaller than those found for the zone boundary of a typical crystal. The conservation of wave vector in scattering by lattice phonons requires that

$$\underline{k}_{0} = \underline{k}_{s} + \underline{q}$$

for phonon creation, where \underline{k}_{0} , \underline{k}_{s} , and \underline{q} are the wave vectors of the incident photon, the scattered photon, and the emitted phonon, respectively. Since, on assuming little dispersion of the refractive index of the medium, $|\underline{k}_{0}|$ is approximately equal to $|\underline{k}_{s}|$, conservation of wave vector leads to the result [24] that

 $q = |q| = 2k \sin \theta$,

where $k_0 = |k_0|$ and θ is the angle between \underline{k}_0 and \underline{k}_s . The physical importance of equation (2) for the present discussion is the fact that $2k_0$ is the approximate magnitude of the <u>maximum</u> phonon wave vector allowed in Brillouin scattering. Since, for usual crystal lattices, (π/a) is much larger than the wave vector k_0 of the incident photon, Brillouin scattering involves only phonons with small values of $q \approx 0$ near the zone center. However, for the electron lattice, (π/a) is roughly equal to k_0 for visible and near infrared photons, so it is to be

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expected that LA and TA vibrational modes with wave vectors q of magnitude equal to the zone edge values would take part in the Brillouin scattering process.

It appears reasonable, then, to conclude that the Brillouin scattering spectrum (i.e., intensity as a function of photon frequency shift $\Delta \omega$ for a given scattering direction θ) of the electron lattice should show new features in addition to scattering by the LA (k = 0) mode at $\Delta \omega = \omega_{\rm p}$ found for the electron gas. Since both the LA and TA vibrations with wave vectors covering, approximately, the whole Brillouin Zone would be expected to take part, spectral structure at values of $\Delta \omega$ smaller than $\omega_{\rm p}$ is to be anticipated. The relative intensities will depend, among other things, on the occupation numbers of the various phonon modes of the electron lattice and, hence, on the temperature. It is also to be expected that ω_{ϱ} and ω_{μ} would, through their proportionality to ω_{p} , be directly proportional to the square root of the electron density n. This behavior, which has been observed [25] for Brillouin scattering by plasmons in the electron gas, would serve to identify observed scattering peaks as due to scattering by vibrations of the electron lattice. In any event, it appears, based on the ideas presented above, reasonable to expect significant qualitative differences between the Brillouin scattering spectrum due to plasmons in the electron gas and the features anticipated for the spectrum of the electron lattice.

A calculation of the Fermi-Thomas screening length $\lambda_{\rm FT} \equiv (6\pi {\rm ne}^2/\epsilon {\rm E}_{\rm F})^{-1/2}$, where ${\rm E}_{\rm F}$ is the Fermi energy, for an electron density n = 10^{14} cm⁻³ in germanium yields a value of $\lambda_{FT} = 200$ Å. Further, one would expect to use an incident photon frequency ω_0 for scattering experiments such that

$$\omega_{\rm p} < \omega_{\rm o} < (E_{\rm G}/\hbar)$$

where E_{G} is the energy gap of the semiconductor in question. For the germanium described above $\omega_{p} = 4.1 \times 10^{11} \text{sec}^{-1}$ and $(E_{G}/h) =$ $1.14 \times 10^{15} \text{sec}^{-1}$. The corresponding wavelengths $\lambda_{p} = (2\pi c/\omega_{p}) =$ 0.463 cm and $\lambda_{G} = 2\pi c/(E_{G}/h) = 1.654 \times 10^{-4} \text{ cm}$. The incident photon wavelength λ_{o} used will then be such that it is less than λ_{p} and greater than λ_{G} , so it will then be true that $\lambda_{o} \geq \lambda_{FT}$. This means that the electron gas at this density should exhibit collective (i.e., plasmon) behavior, rather than single particle behavior, for an incident wavelength λ_{o} in the Brillouin scattering experiments suggested above.

It is therefore reasonable to suggest that an experimental study of Brillouin scattering in very pure germanium (doped with antimony) would be a reasonable method of searching for the Wigner lattice. A series of such samples with varying values of the donor atom density N_d could be studied at appropriate temperatures, yielding a series of Brillouin scattering spectra at varying values of the electron density n in a given temperature. A search for the spectral features described above would then probe the presence or absence of the electron lattice.

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IV. Stability of the Lattice

In order to investigate the potential stability of the Wigner lattice in the temperature range in question, the magnitude and sign of the cohesive energy may be calculated. The cohesive energy ΔE , per electron, of the lattice is the difference between the energy E_g of the electron gas per electron and the energy E_s of the crystal per electron:

$$\Delta E \equiv (E_g - E_s).$$

A positive value of ΔE will imply the stability of the lattice relative to the gas for the value of the electron density n being considered. The energy E_s at T = 0 K, when the vibrations of the lattice have the average phonon occupation number $\langle n \rangle = 0$, is given [4] by

$$E_s = [(-3e^2/2r_o) + (1/2)\sqrt{3} \hbar \omega_p],$$

and the energy E_{g} may be taken as (3/5) E_{F} , where E_{F} is the Fermi energy at the electron density n. The expression for E_{s} may be modified to

$$s = [(-3e^2/2r_0) + (\langle n \rangle + 1/2)\sqrt{3} \hbar \omega_p]$$

to include the case of values of $\langle n \rangle$ not equal to zero.

If we consider T = 16K, $k_B T = 1.38 \times 10^{-3} eV$, and, for n = $10^{14} cm^{-3}$, $\hbar \omega_p = 2.69 \times 10^{-4} eV$, so $k_B T > \hbar \omega_p$, leading to the result that

$$\langle n \rangle \cong (k_{\rm B}T/\hbar\omega_{\rm p}) \cong 5.$$

Using <n> = 5 and $r_o = (3/4\pi)^{1/3} n^{-1/3} = 1.34 \times 10^{-5}$ cm, one obtains $E_s = -2.16 \times 10^{-14}$ ergs per electron, $E_g = 0.624 \times 10^{-16}$ ergs per electron, and ΔE is found to be equal to 2.17 x 10^{-14} ergs per electron or 1.36 x 10^{-2} eV per electron. The cohesive energy is thus positive and its value may be compared to the cohesive energy of solid neon (melting point 24K) of 0.02 eV per atom given in Reference 11, page 99. We see also that $(\Delta E/\hbar\omega_p) \approx 50$ and $(\Delta E/k_BT) \approx 10$, showing that the cohesive energy per electron of the Wigner lattice is large, in this temperature range, compared to both k_BT and the largest quantum $\hbar\omega_p$ of the vibrational spectrum of the lattice. It is therefore reasonable to conclude that the electron lattice is, at least in principle, stable in this temperature range.

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V. Other Possible Experiments

There are, of course, other possible experiments to consider for the detection of the electron lattice. Among these is a study of the specific heat C_{v} as a function of electron density. It appears, however, that the Debye temperature [26] for the electron lattice is about 3000K, while that of the host germanium is 374K. The lattice specific heat of the germanium would thus be a factor of about 10^3 larger than that of the electron crystal, making experimental detection of the latter difficult. One might consider observing the vanishing of the term in γT in the specific heat [27] as the electron gas crystallizes. Howver, a calculation (by the author) of γ for an electron gas of density n = 10^{14} cm⁻³ leads to a value of $\gamma = 9 \times 10^{-2}$ erg-deg⁻². The electron gas specific heat γT is equal to 1.5 erg-deg⁻¹ at T = 16K, compared to a lattice specific heat of 1.1×10^5 erg-deg for germanium, both values being for a volume of 1 cm^3 . It appears, therefore, that experimentally detecting the electron lattice using specific heat measurements would probably be difficult. Another type of possible experiment might involve the scattering [28] of phonons by the conduction electrons in a solid. One might anticipate that the ordered electron lattice would scatter phonons less than does the electron gas, and that this difference might be probed with thermal conductivity measurements, or perhaps with an experiment involving the scattering by electrons of monochromatic phonons generated by the methods used by Dynes and Narayanamurti [29].

VI. Conclusion

In summary, it is proposed that conditions under which the three dimensional Wigner electron lattice is predicted to form may be achieved in extrinsic germanium at low temperatures. Of several possible experiments which might detect this lattice, it appears that the most feasible is the study of Brillouin scattering of photons by the LA and TA modes of vibration of the electron lattice.

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Acknowledgement

The assistance of the U.S.E.R.D.A. is gratefully acknowledged. The author would also like to thank N. Amer, A. F. Kip, A. M. Karo, and A. M. Portis for valuable discussions.

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 10. This value of m* is an average over the longitudinal and transverse masses m_l = 1.64m and m_t = 0.082m [see, for example, R. Dalven, Infrared Phys. <u>6</u>, 129 (1969)] using the relation

 $m^* = 3m_{+}m_{\varrho}/(2m_{\varrho} + m_{+}).$

 C. Kittel, "Introduction to Solid State Physics", Fourth Edition, (John Wiley, 1971), page 374, Table 3. -19-

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- 12. C. Kittel, Reference 11, page 376, equation (16). The requisite condition that $E_d = k_B T$ is satisfied for the cases considered in this paper.
- 13. The decrease of E_d with increasing donor concentration N_d for values of N_d larger than about 10¹⁵ cm⁻³ [see, for example, S. Wang, "Solid State Electronics", (McGraw-Hill, 1966), page 157] has been neglected for simplicity.
- 14. A second possible semiconductor for this purpose is GaAs. Even though GaAs has a simpler conduction band structure than does germanium, the present technology of GaAs is not as advanced as is that of germanium, and it is doubtful that GaAs of purity sufficient for these purposes is available.
- 15. P. M. Platzman and P. A. Wolff, Reference 3, page 14 and 85.
 16. By a "simple semiconductor" is meant a semiconductor with a single, isotropic conduction band minimum. Examples are GaAs (see Note 14) and InSb. Germanium, with its many-valley conduction band, is not member of this class, but the complications thereby introduced (see Reference 3, pages 99ff) are ignored in this paper for simplicity.
- 17. For a review, see P. M. Platzman and P. A. Wolff, Reference 3, Section 34, pages 85ff.
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21. In the example under discussion, this uniform background of positive charge is the randomly positioned ionized donors in the germanium crystal. It may be (A. M. Portis, private communication) that the localized nature of the donors and electrostatic screening will make them a poor approximation to the <u>uniform</u> positive background assumed in theoretical studies of the Wigner lattice. A better approximation might be the intrinsic holes produced (along with electrons) by say, band gap photons. However, the scheme proposed in the text appears at least a reasonable way to achieve experimentally an approximation to the Wigner conditions.

22. W. A. Harrison, "Solid State Theory", (McGraw-Hill, 1970), page 288.

- 23. See, for example, G. F. Koster, Solid State Physics, (F. Seitz and D. Turnbull, editors), <u>5</u>, page 25, Fig. 6 (Academic Press, 1957).
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- 25. See Reference 3, page 86.
- 26. Calculated by the author, using $r_s = 19$, from the relation given by Carr on page 1444 of his paper in Reference 6. This value of the Debye temperature of the electron lattice must, however, be regarded with caution since it corresponds to the very high value of 2 x 10⁹ cm/sec for the velocity of sound.
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