Laser Doppler Velocimetry of Electron-Hole Drops in Germanium

J. Doehler, J. C. V. Mattos,* and J. M. Worlock Bell Telephone Laboratories, Holmdel, New Jersey 07733 (Received 7 February 1977)

We report the first direct measurement of velocity distributions of electron-hole drops, and discuss them in relation to existing theoretical models. The data, obtained by frequency analysis of scattered light, show acceleration near the surface, while at larger depths, the drops appear to experience viscous deceleration with a relaxation time of 10^{-4} sec. We find difficulties in reconciling the data with the present ideas of forces resulting from excitons, phonons, or mutual repulsion.

While the structure of electron-hole drops (EHD) in Ge is now reasonably well understood,¹ many facts, such as those associated with their motion, remain unknown. Several experiments have been performed to measure EHD velocities. These experiments are primarily based on measuring the time of arrival of droplets at some point in the crystal remote from the point of pulsed optical injection. Results interpreted in terms of rectilinear motion²⁻⁵ yield velocities always decreasing with distance and time, sometimes initially exceeding the sound velocity. Results interpreted in terms of translational diffusion yield widely ranging values for the EHD diffusion constant D, from 10^{-3} to 500 cm²/sec.^{2,6-9} In this Letter, we report the first direct measurement of EHD velocities, and demonstrate a technique which will be useful for measuring such velocities under many different experimental conditions.

The setup is shown in Fig. 1. The sample was of high-purity Ge ($|N_A - N_D| < 3 \times 10^{10} \text{ cm}^{-3}$) obtained from GE Company. The geometry and preparation of the sample were similar to those reported in Ref. 4. It was immersed in superfluid helium maintained at 2 K. EHD were produced by focusing the beam from an argon-ion laser ($\lambda = 5145$ Å) to a point approximately 200 μ m in diameter on a Syton-polished face. The probe light, obtained from a He-Ne laser ($\lambda = 3.39$ μ m), was scattered by the EHD, and collected at an angle controlled by the position of mirror M_{2} along an axis perpendicular to the probe beam. Heterodyne detection was performed by mixing the scattered light, whose spatial coherence was assured by diaphragm D_1 , with a reference beam obtained by beam splitting the incident light. The beat frequencies were analyzed with a Hewlett-Packard 141T spectrum analyzer whose bandwidth was held constant at 3 kHz and whose output was squared to obtain proper power spectra and to

achieve proper noise subtraction in the lockin amplifier. The detector consisted of a liquidnitrogen-cooled HgCdTe chip having a 250-nsec time constant.

Light scattered by a particle moving with a rectilinear velocity $\vec{\mathbf{v}}$ is Doppler-shifted in frequency by an amount given by $\Delta f = (2\pi)^{-1}(\vec{\mathbf{v}}\cdot\Delta\vec{\mathbf{k}})$, where $\Delta\vec{\mathbf{k}}$ is the change in photon wave vector.¹⁰ In contrast, the spectrum of light scattered by particles of mass *m* undergoing diffusional motion with a diffusion constant $D = k_{\rm B}T\tau/m$, governed by a velocity relaxation time τ , is a Lorentzian centered at $\Delta f = 0$, whose width is given



FIG. 1. Schematic outline of the Doppler velocimeter used in this experiment. P is a quartz quarter-wave plate used for optical isolation; the incident beam was thus circularly polarized at the sample. The mechanical arrangement was such that the sample could be accurately positioned and/or replaced with a piece of ground glass used for alignment. The drawing is not to scale.



FIG. 2. (a) Raw scattered light spectrum (noisy line) and spectrum corrected for the detector/amplifier frequency response (dashed line). (b) Mean frequency shift vs scattering angle (inside the sample). Pump power and probing depth were held constant at 55 mW and 330 μ m, respectively. The open circle indicates datum point obtained from (a). The slope of the straight line is thus proportional to the average EHD velocity. The deviation at high angles is not significant.

by $\Gamma_D = (2\pi)^{-1}D |\Delta \vec{k}|^2$. Note that Δf depends linearly on scattering angle θ (for small θ), while Γ_D depends on θ^2 .

Figure 2(a) shows a typical spectrum recorded in our experiment, while Fig. 2(b) shows that the dependence of the mean frequency shift on scattering angle is linear. We find that the width and the position of the finite frequency peak in Fig. 2(a) follow the same linear angular dependence. This important result shows immediately that the motion of the drops is observed to be rectilinear rather than diffusive and allows the construction of the velocity scale drawn on top of Fig. 2. A necessary condition for the observation of diffusional motion is that $\Gamma_D \tau \ll 1$. We thus conclude that this condition is violated (i.e., τ is larger than the inverse of a typical frequency shift shown in Fig. 2, and $D \ge 0.7 \text{ cm}^2/\text{sec}$) or that the diffusional broadening is overwhelmed by a spread in the velocity distribution shown in Fig. 2 (i.e., Γ_p $< 10^5$ Hz, and D < 0.05 cm²/sec).

The average velocities, obtained from the mean



FIG. 3. Average EHD velocity, obtained from mean frequency shifts, as a function of probing depth and pump power. The lines, connecting data points taken with constant pump power, have been drawn for clarity. The temperature and scattering angle were held constant at 2.0 K and 0.03 rad, respectively.

frequency shifts, are plotted in Fig. 3, as a function of depth for various pump powers. The EHD are seen to accelerate, essentially from rest, up to a depth of about 250 μ m; the acceleration rates, and consequently the maximum velocities, are power-dependent. Note that a linear increase of velocity with depth implies a similar linear increase of the particle acceleration. At larger depths, the EHD appear to experience a viscous drag with a relaxation time constant of (1 ± 0.2) $\times 10^{-4}$ sec, independent of pump power. In the following, we discuss the droplet motion in terms of three models, taking the mean velocity to be the EHD velocity, with the idea that each drop follows a complete trajectory from the surface to the crystal interior.

(1) "Exciton wind" model. —In this model, EHD are accelerated and decelerated by momentum exchange with free excitons (FE). At large depths, the FE are assumed to be at (constant) vapor pressure density and stationary with respect to the lattice. This atmosphere damps the ballistic motion of the drops. Our measured relaxation time of 10^{-4} sec agrees well with estimates by Combescot¹¹ and requires FE densities of the order of 10^{12} cm⁻³. Near the surface, EHD are accelerated by the "exciton wind" as first proposed by Balslev and Hvam.¹² Our observed acceleration rates require reasonable FE density gradients of the order of 10^{13} cm⁻⁴. With a relaxation time of 10^{-4} sec, the diffusion constant becomes of the order of 25 cm²/sec, which lies outside of our accessible range. There are three major objections to this model. First, since the acceleration increases with depth in the first 200 μ m, the FE density gradient must also increase, and this requires a very peculiar density distribution. Second, there is no evidence in our spectra of an approach to thermal velocity distribution at large depths even though the EHD slow to characteristic velocities below the thermal velocity of about 500 cm/sec. Third, it ignores the interaction of EHD with phonons, which is discussed next.

(2) "Phonon wind" model.—Absorption, scattering, and emission of thermal phonons by EHD provide a viscous damping of their motion, while their interaction with a flux of nonthermal phonons provides an accelerating force. Theoreti cal^{13} and experimental^{6,7,14} studies of these effects lead us to believe that the EHD momentum relaxation time can be very short ($\tau \leq 10^{-8}$ sec). In this case, diffusional motion is very slight (D < $< 10^{-3} \text{ cm}^2/\text{sec}$) and again outside of our measurable range. With such a short relaxation time, the observed (steady-state) velocities are proportional to the force on the droplet. In turn, this force is proportional to the flux of nonthermal phonons, whose greatest source is the excitation process. About 1.7 eV of excess energy per e-hpair is carried away from the excited surface in the form of phonons. Following Keldysh,¹⁵ we can calculate that there is potentially 100 times the phonon flux required to generate our observed velocities. However, only phonons of wave vector less than twice the Fermi wave vector are effective; a model which deals with the conversion of high-frequency quanta into effective long-wavelength phonons may perhaps explain the numerical discrepancy as well as the initial increase of the pushing force with depth.

(3) Hydrodynamical model.—Here, strong EHD-EHD interaction govern the EHD motion. Such an interaction could be provided, for instance, by the electrostatic charge that the EHD have been predicted¹⁶ and shown¹⁷ to possess, or by an exchange of phonons.¹⁵ However, difficulties arise in attempting to explain the behavior near the surface, and an additional mechanism, such as phonon or exciton damping, is required to account for the decreasing velocities at larger depths.

In conclusion, none of the above models is able to account adequately for our measured velocities. We have, however, demonstrated a technique for measurement of EHD velocities. It is clear that these velocities are sensitive to interaction with other particles, such as excitons, phonons, and photons; to impurities and other drops; as well as to externally applied fields such as electric, magnetic, or strain fields. Experiments probing such droplet interactions should prove interesting and informative. In particular, we hope to investigate the interaction with phonons in the near future by (a) varying the wavelength of the exciting radiation, which will change the number of phonons generated in the excitation process, and (b) inserting well-characterized, independently generated heat pulses.

We gratefully acknowledge valuable discussions with J. P. Gordon, T. M. Rice, M. Combescot, and R. E. Slusher, and the expert sample preparation of A. A. Pritchard and A. L. Albert. J. C. V. Mattos acknowledges the generous support of the Foundacão de Amparo a Pesquisa do Estado de São Paulo.

*Supported by a Foundacão de Amparo a Pesquisa do Estado de São Paulo Fellowship. Permanent address: Universidade Estadual de Campinas, Instituto de Fisica, 13100 Campinas, São Paulo, Brazil.

¹For a recent review, see, for example, M. Voos and C. Benoît à la Guillaume, in *Optical Properties of Solids, New Developments*, edited by B. O. Seraphin (North-Holland, Amsterdam, 1976), p. 143.

²C. Benoît à la Guillaume, M. Voos, and F. Salvan, Phys. Rev. Lett. <u>27</u>, 1214 (1971).

³A. S. Alekseev, V. S. Bagaev, and T. I. Galkina, Zh. Eksp. Teor. Fiz. <u>63</u>, 1020 (1973) [Sov. Phys. JETP <u>36</u>, 536 (1973)].

⁴T. C. Damen and J. M. Worlock, in *Proceedings of* the Third International Conference on Light Scattering in Solids, Campinas, Brazil, 1975, edited by M. Balkanski, R. C. C. Leite, and S. P. S. Porto (Wiley, New York, 1976).

 $^5 J.$ M. Hvam and I. Balslev, Phys. Rev. B $\underline{11}, 5053$ (1975).

⁶A. S. Alekseev et al., in Proceedings of the Twelfth International Conference on the Physics of Semiconductors, Stuttgart, Germany, 1974, edited by M. H. Pilkuhn (B. G. Teubner, Stuttgart, Germany, 1974), p. 91.

⁷J. C. Hensel and T. G. Phillips, in *Proceedings of* the Twelfth International Conference on the Physics of Semiconductors, Stuttgart, Germany, 1974, edited by M. H. Pilkuhn (B. G. Teubner, Stuttgart, Germany, 1974), p. 51.

⁸Y. E. Pokrovskii, Phys. Status Solidi (a) <u>11</u>, 385 (1972).

⁹R. W. Martin, Phys. Status Solidi (b) <u>61</u>, 223 (1974). ¹⁰For a discussion of quasielastic light scattering, see, for example, H. Z. Cummins and H. L. Swinney, in *Progress in Optics*, edited by E. Wolf (North-Holland, Amsterdam, 1970), Vol. 8.

¹¹Monique Combescot, Phys. Rev. B 12, 1591 (1975).

¹²I. Balslev and J. M. Hvam, Phys. Status Solidi (b) <u>65</u>, 531 (1974).

¹³L. V. Keldysh and S. G. Tikhodeev, Pis'ma Zh. Eksp. Teor. Fiz. <u>21</u>, 582 (1975) [JETP Lett. <u>21</u>, 273 (1975)].

¹⁴A. S. Alekseev *et al.*, Pis'ma Zh. Eksp. Teor. Fiz. <u>21</u>, 578 (1975) [JETP Lett. <u>21</u>, 271 (1975)]; B. Etienne, L. M. Sander, C. Benoît à la Guillaume, M. Voos, and J. Y. Prieur, Phys. Rev. Lett. 37, 1299 (1976).

¹⁵L. V. Keldysh, Pis'ma Zh. Eksp. Teor. Fiz. <u>23</u>, 100 (1976) [JETP Lett. 23, 86 (1976)].

¹⁶For the most recent calculation of droplet charge, see R. K. Kalia and P. Vashishta, to be published. ¹⁷Y. E. Pokrovskii and K. I. Svistunova, Pis'ma Zh. Eksp. Teor. Fiz. <u>19</u>, 92 (1974) [JETP Lett. <u>19</u>, 56 (1974)].

Have We Seen a Heavy Antinucleus?*

Ray Hagstrom

Lawrence Berkeley Laboratory, University of California, Berkeley, California 94720 (Received 3 January 1977)

The hypothesis of the first observed heavy antinucleus is proposed for the event of Price *et al*. This hypothesis demands depletion of the high-energy knock-on electrons, which should be clearly evident in the nuclear emulsion. This hypothesis does not directly contradict any previous antimatter search; and conflict with extrapolations from previous searches is of small statistical significance.

The cosmic-ray event of Price *et al.*¹ has received much critical comment. Several authors have proposed alternative hypotheses to the originally claimed identification.²⁻⁵ Experimental interpretation of this event will be briefly discussed and a specific experimental recommendation will be made to help settle the experimental controversy. I will furthermore propose that the experimental descriptions of the particle to date suggest identification as an antinucleus. Quantitative predictions must be matched by the nuclear emulsion data to substantiate the antinucleus hypothesis. The cosmological consequences of the observation of a heavy antinucleus would be considerable. Let us consider, in turn, the results from each of the detectors.

As nuclei slow, their ionization rises steeply. This particular particle had so high an ionization rate $(|Z/\beta| \approx 114)$ that, were it any nucleus previously seen among the cosmic rays, it should have slowed measurably in passage through the 0.9-g/cm^2 Lexan stack.¹ For instance, a uranium nucleus would be expected to have its Z/β increased by three units.¹ Nevertheless, its ionization did not rise.¹ This particle appears to be the most penetrating particle with such a high ionization seen to date.¹ However, interpretation of this observation has led to controversy.

It is agreed that the Lexan data cannot be reasonably reconciled with any slow ($\beta \le 0.6$) normal nucleus, fragmenting or otherwise.^{1,4,5} Given a possible, but unlikely, chain of fragmentations, normal nuclei with speeds $\beta > 0.65$ have been con-

sidered to fit the Lexan data.¹⁻⁵ The Lexan data better match nuclei with higher speeds so that, for fast normal nuclei ($\beta > 0.8$), agreement is reasonably within experimental tolerances.¹⁻⁵

It has long been known that negative particles are more penetrating than their charge conjugates.⁶ The difference is due to higher-order electrodynamics. It has not been universally realized how large these corrections can be for heavy nuclei. Using order- Z^3 calculation for the distant collisions⁷ (which is a tiny correction) together with an exact form for the close collisions,⁸ I find that the stopping powers for nuclei and their charge conjugates differ by 15% to 25% in the appropriate realm. These differences arise predominantly from close collisions. Thus, we should expect the heaviest antinuclei to be the most penetrating particles of any given ionization seen to date. Because Lexan responds predominantly to distant collisions,⁹ we expect its response to reflect $|Z/\beta|$ independently of the sign of Z.

It has been pointed out, for instance in Refs. 2– 5, that reactions which noncataclysmically diminish $|Z_{\text{projectile}}|$ allow the projectile to appear more penetrating. Such interactions are featured in normal-nucleus explanations of the event with $0.65 \le \beta \le 0.80.^{1-5}$ One wonders whether such processes might occur when antinuclei penetrate the Lexan. I have no cross sections for specific channels of charge loss in collisions of antinuclei with normal nuclei excepting (\bar{p}, p) and (\bar{p}, d) . In the appropriate speed range, the (\bar{p}, p) cross sec-