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EFFECT OF PHONON FLUX SINGULARITIES ON THE CLOUD OF ELECTRON-HOLE DROPLETS

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A comparison is made between the sharp structure observed in the cloud of electron-hole droplets in Ge and the phonon flux emanating from a point source.

Generally, the low temperature photo-excited phases of Ge and Si are produced by surface excitation with photon energies well above the band gap. In high purity crystals the free excitons (FE), having relatively light mass $m=m_e+m_h$, can migrate deep into the crystal (> 1mm) before recombining. This is a consequence of their high mobility at low temperature and long lifetime due to the indirect band edge. On the other hand, electron-hole droplets (EHD) are much less mobile than FE, by virtue of their much larger mass ($\sim 10^3$m in Si and $\sim 10^7$m in Ge). Even so, EHD are also observed to travel about a millimeter before decaying. This is because droplets are readily pushed by nonthermal phonons, such as those generated in the initial thermalization of the hot photoexcited carriers. The force on a droplet arises from the absorption of the phonon momentum $\hbar\vec{q}$, as originally proposed by Keldysh [1]. The study of this phonon-wind drift process in the past several years has provided an interesting contrast to the diffusive motion of FE, and has revealed a complex but fascinating interaction between phonons and the electron-hole liquid.

While diffusion of particles in a crystal of cubic symmetry would produce an isotropic distribution of particles, a phonon-wind drift process can lead to large anisotropies. This was experimentally demonstrated in Ge by Greenstein and Wolfe [2], who observed by luminescence imaging that the cloud of electron-hole droplets emanating from a point excitation was extremely non-spherical. The features of this cloud were qualitatively explained in terms of a) the anisotropic electron-phonon coupling and b) the anisotropic flux of phonons from a point source--a phenomenon known as phonon focusing. These experiments showed that a significant component of the phonon wind was emanating near the excitation point, probably as a byproduct of the rapid thermalization of hot carriers. Since the initial results we have developed two refined imaging techniques to characterize 1) the angular-dependent transport of <u>droplets</u> and 2) the anisotropic energy flux of <u>phonons</u> emanating from a point source.

The first technique involves a computer-controlled imaging system which produces constant density contours of the electron-hole droplet cloud as a function of distance from the excitation point. The second method, which we call ballistic phonon imaging, produces a two-dimensional map of the phonon flux itself. Together, these two experiments show graphically the intrinsic role of non-thermal phonons in transporting EHD. In this paper we give the first experimental comparison of the electron-hole droplet distribution with the two-dimensional angular distribution of phonons emanating from a laser-produced heat pulse. The influence of phonon focusing on the cloud shape is both striking and unequivocal. The sharp features of the cloud can now be readily explained, and some interesting new details come to light which were not anticipated before the ballistic phonon imaging method was introduced.

Figure (1) shows the orientation of a $1cm^3$ ultrapure dislocation-free crystal of Ge used in both experiments and typically cooled to 1.8K. The crystal is photoexcited by a continuous YAℓG:Nd laser beam, focused to a 100 μm spot size. An image of the resulting cloud of electron-hole droplets is obtained by collecting the droplet luminescence through the $(00\bar{1})$ surface, as shown in Fig. (2a). This image is recorded digitally as described elsewhere [3]. Fig. (2b) is a contour map of the image showing more accurately the shape of the droplet cloud. The cloud is characterized by broad lobes and sharp flares, and the penetration depth of droplets into the sample increases with excitation power. The basic interpretation of this structure [2,4] is that a rather localized source of ballistic phonons, with intensity proportional to laser power, pushes droplets from the excitation region, where they are created, deep into the crystal. The localized nature of this phonon source is apparent from the sharpness of the flares and the observation in Fig. (2) that they emanate directly from the excitation point.

An explanation for these sharp structures in the cloud lies in the basic anisotropy of ballistic phonon flux emanating from a point source of heat. Taylor, Maris, and Elbaum [5] originally demonstrated this effect in crystals, and more recent calculations by Rösch and Weis [6] and experiments by Hensel and Dynes [7,8] have shown the existence of sharp features in the TA phonon flux in Ge. Most recently, Northrop and Wolfe [9] have developed a method for imaging the phonon flux radiating from a point heat-pulse. This method and its theoretical interpretation provide a new geometric visualization of the anisotropic propagation of phonons in crystals.

The ballistic phonon intensity emanating from YAℓG:Nd photoexcitation of Ge is graphically shown in Fig. (3a). The bright regions correspond to directions of intense TA phonon flux. The images are in fact obtained by scanning a laser-produced heat-pulse across three surfaces of the crystal and detecting the phonon intensity with a superconducting bolometer at the center of the (110) face. The sharp features actually correspond to mathematical infinities in the phonon flux, as described in [9]. Using the known elastic constants of Ge, these computed singularities are plotted in Fig. (3b) for the slow and fast TA modes. Agreement between theory and experiment is excellent.

We can now see the origin of the sharp flares in the electron-hole droplet cloud. The fast TA mode produces {100} planes of high phonon flux. These planes--two of which are viewed edgewise in Fig. (2a)--provide the main contribution to the flare structure. Small rotations of the crystal are found to produce a marked broadening of the flares, verifying the planar nature of these structures. By viewing the cloud in other orientations the effect of the concentrated slow-TA-flux near the <100> axes can also be observed. Thus we can conclude that the sharp flares in the cloud have both a planar and axial character, consistent with the heat-pulse images.

While the longitudinal phonons have no flux singularities, their

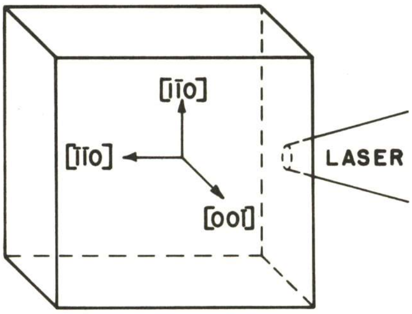


Fig. (1) Experimental configuration for the infrared imaging experiment: A continuous, focused laser beam produces a cloud of EHD. Thermalization phonons are also produced near the excitation point which push droplets into the crystal

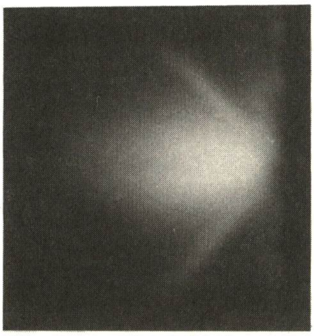
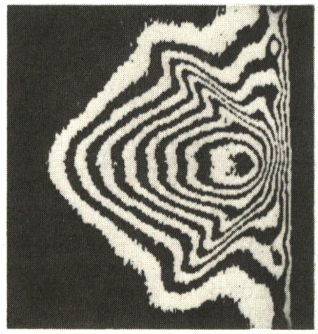
(a)  (b) 

Fig. (2a) Cloud of electron-hole droplets as viewed along the excitation surface (vertical line) and detected by the recombination luminescence at 1.75 μm: A broad lobe protrudes along the <111> axes and sharp flares "apparently" along $[\bar{1}00]$ and $[0\bar{1}0]$ axes are prominent. Photo width = 1.6 mm. Laser power = 15 mW
(b) Contours of constant luminescence intensity for the image of Fig. (2a)

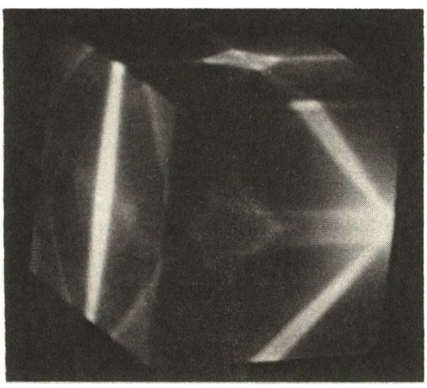
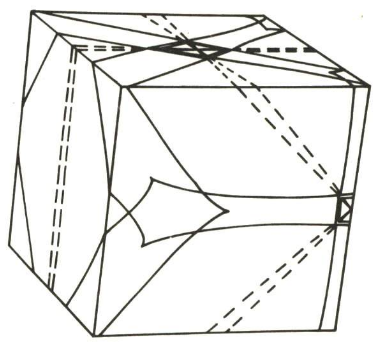
(a)  (b) 

Fig. (3a) Ballistic phonon image for a crystal of Ge oriented as in Fig. (1): The bright regions are directions of high transverse-acoustic phonon flux and may be viewed as emanating from a point at the laser spot of Fig. (1). The intense ridge of intensities in the (100) and (010) crystal planes provide a large phonon wind force which creates the flare structure in the EHD cloud of Fig. (2)
(b) Theoretical calculation of the phonon flux singularities for the geometry of Fig. (3a): Dashed lines are for fast TA phonons and solid lines are for slow TA phonons

intensity is broadly concentrated along <111> [5,6,7]. Thus, the effect of longitudinal phonons on the cloud is to produce broad lobes along the <111> directions. A quantitative calculation of these effects which includes the anisotropic electron-phonon coupling has been carried out by Markiewicz et. al. [10] and shows good agreement with the measured contour maps. Additional features considered in these calculations include the attenuation of the phonon wind by droplet absorption and the occurrence of nonradial droplet trajectories. Further experiments and calculations are now underway to understand the details of the droplet distribution in Ge and also in Si [11].

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