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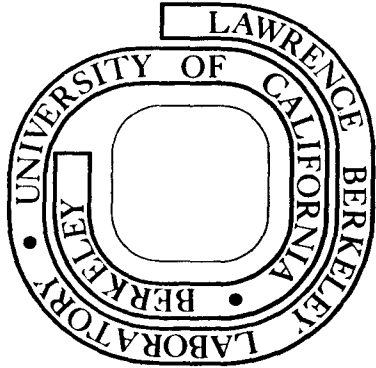
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DISCOVERY OF THE IMMOBILITY OF ELECTRON-HOLE

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DISCOVERY OF THE IMMOBILITY OF ELECTRON-HOLE DROPS IN Ge AT LOW EXCITATION

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Abstract

Using a new experimental technique based on the hysteresis effect, the spatial distribution of a localized packet of electron-hole drops in a spatially uniform free exciton gas near threshold is measured for times $\sim 10^4$ sec. No significant motion is found, determining an upper limit $D \lesssim 10^{-9}$ cm^{2}sec^{-1} to the drop diffusion constant. These results require that drops bind to crystal defects; for electrical impurities we estimate the binding energy $B \gtrsim 5$ meV.

Mobility is a fundamental property of electron-hole drops (EHD) and the motion of EHD in Ge has been extensively studied in past years.[1] The motion of drops has been commonly assumed to be diffusive, and early measurements[2,3] of the spatial distribution of drops gave values for the EHD diffusion constant $D \simeq 0.1$ cm^{2}sec^{-1} to $D \simeq 500$ cm^{2}sec^{-1}. The tremendous variation in these early experimental results is now believed to be a consequence of the dynamic nature of the EHD "cloud"[1] produced in these experiments. Drops are propelled from the excited region by forces,[4] notably the phonon wind,[5] due to the high excitation levels commonly used. These EHD travel into the crystal and eventually decay, forming a dynamic cloud of drops. In other work[6] the value $D \sim 10^{-3}$ cm^{2}sec^{-1} was indirectly obtained from measurements of the mobility of EHD in Ge under non-uniform deformation. All of the above experiments represent the motion of EHD in the presence of perturbing forces.

In this paper we present a sensitive new experimental technique which measures the intrinsic motion of EHD in a spatially-uniform free exciton (FE) gas at very low excitation, in which the forces on EHD are negligible. The experimental method is based upon the creation of a localized packet of electron-hole drops in a spatially-uniform FE gas, using the phenomenon of hysteresis.[7] The very slow time development of the resulting spatial distribution of EHD is then obtained by repeatedly measuring the spatial profile of the EHD luminescence. We find that EHD are essentially immobile in ultrapure Ge, and measure the upper limit $D \lesssim 10^{-9}$ cm^{2}sec^{-1} to the EHD diffusion constant at 2.1 K, a value 6 to 11 orders of magnitude smaller than previous

estimates.[2,3,6] We conclude that EHD are pinned to crystal defects

in the absence of perturbing force.

A sketch of the Ge sample to scale, showing the excitation and

luminescence collection geometry is given in Fig. 1a. Data from two

large crystals of ultrapure[8] Ge are reported here: sample 145M

($12 \times 12 \times 5$ mm^3), which is undislocated with net impurity concen-

tration ($N_A - N_D) \simeq 2 \times 10^{11}$ cm^{-3} primarily due to 80 meV acceptor

levels,[9] and sample 437D ($8 \times 8 \times 4$ mm^3), which is dislocated ($N_{dis} \sim$

~ 500 cm^{-2}) with net impurity concentration ($N_A - N_D) \simeq 2 \times 10^{10}$

cm^{-3} due to shallow impurities only. The samples were crystallographically

oriented, mechanically cut and polished to an optical finish, and

the excited surface was etched in 10:1 HNO$_3$:HF. The samples were

mounted free of strain or electrical contacts and immersed in pumped

liquid He. As indicated in Fig. 1a, the entire front surface of each

sample was uniformly illuminated with 1.5 μm radiation derived from

a tungsten-halogen lamp with an interference filter; the absorption

length of this wavelength light is ~1 mm, providing volume excitation

of the Ge crystal. Additional surface excitation in a stripe

0.1×7 mm^2, was provided by a focussed Ar$^+$ ion laser as shown.

Luminescence from EHD and FE was collected from the back face of the

samples and spectrally analyzed by a 1/4 m spectrometer and a sensitive

Ge photodetector. As indicated in Fig. 1a, the 0.2 mm wide image

of the spectrometer slit was scanned across the back face of the crystal

by translating the collection lens. Scans of the LA phonon assisted

FE and EHD luminescence intensities I_{FE} and I_{EHD}, schematically indicated

in Figs. 1b and 1c, were obtained using an automated digital signal averaging apparatus.

The experimental procedure is demonstrated by the data from sample 145M given in Fig. 2. Initially the uniform volume excitation is monotonically increased from zero to a constant level P. As indicated on the schematic hysteresis curves in Fig. 1d, the level P is chosen below the threshold P_+ for the creation of EHD, but above the threshold P_- for drop breakup. Thus the presence of EHD, indicated by I_{EHD} in Fig. 1d, depends upon the history of excitation. Initially no drops are present in the sample, as confirmed by scans of I_{FE} and I_{EHD} in Figs. 2a and 2b, and the density of FE is reasonably uniform (the small features at $x \simeq -.5$ mm and $x \simeq -3.0$ mm in Fig. 2 are due to scratches on the back face). A localized packet of EHD is then created by additional momentary stripe excitation. During stripe excitation drops are ejected from the excited region by the relatively large excitation level $P_{stripe} \sim 40$ mWcm^{-1}, as in previous experiments,[1,2] and form the EHD cloud shown in Fig. 2c. After stripe excitation has ceased, the drops come to rest and shrink in size, yielding the EHD spatial distribution given in Fig. 2d. It is essential to realize that the localized packet of EHD shown in Fig. 2d is produced solely as a result of a non-uniform history of excitation. Note also that, in principle, individual drops can survive indefinitely, nourished by the FE gas, at any horizontal position x inside the crystal. From the luminescence intensities I_{FE} and I_{EHD} in Figs. 2a and 2d we estimate that, inside the EHD packet, the system is composed primarily of excitons, and EHD constitute only a small perturbation, unlike previous experiments

on drop motion.[1,2]   The large size of the EHD spatial distributions
shown in Figs. 2c and 2d is characteristic of undislocated Ge, and
the structure is caused by anistropies in the phonon wind[5] and its
interaction with EHD, as discussed by Greenstein and Wolfe.[10]
Sample 145M was oriented such that each face of the crystal was
in a (100) plane.

A series of scans of the EHD luminescence profile at various
times t following the end of stripe excitation are shown in Fig. 3
for sample 145M at 2.10K. The most striking feature of this data
is that the EHD spatial distribution at t $\simeq$ 15,880 sec, Fig. 3c, is
virtually identical to that at t $\simeq$ 130 sec, Fig. 3a. The peak intensity
decreases slightly during this time, but neither the horizontal position
nor the full width at half maximum $\Delta(t)$ vary by more than 30 μm, roughly
the limit of mechanical stability of our apparatus. Data obtained
in an identical manner is shown in Fig. 4 for sample 437D at 2.10K;
note the expanded horizontal scale relative to Fig. 3. As above,
the EHD spatial distributions shown in Fig. 4 are virtually fixed
up to t $\simeq$ 16,600 sec; neither the horizontal position nor the width
$\Delta(t)$ varies by more than 20 μm over this time. The smaller width
and lack of structure of the EHD cloud in Fig. 4 is characteristic
of dislocated Ge.

If electron-hole drops undergo diffusion, the EHD spatial distribution
broadens in time. By solving the one-dimensional diffusion equation
$\partial N/\partial t = D\partial^2 N/\partial x^2$ for the drop concentration $N(x,t)$, we find

$$N(x,t) = N_0 (4\pi Dt + 2\pi\sigma^2)^{-1/2} \exp\left[-x^2/(4Dt + 2\sigma^2)\right] \qquad (1)$$

where the initial distribution $N(x,0)$ is approximated by a Gaussian of variance σ^2. Roughly equating $N(x,t)$ with the measured luminescence profile $I_{EHD}(x,t)$, we expect the full width at half maximum $\Delta(t)$ of I_{EHD} to increase as

$$\Delta(t)^2 = \Delta(0)^2 + (16 \ln 2) Dt \qquad (2)$$

where the finite spatial resolution of the apparatus is included in $\Delta(0)$. Using Eq. (2), we find from the data of Fig. 3 the upper limit $D \lesssim 10^{-8}$ cm^2 sec^{-1} to the EHD diffusion constant at 2.1K for undislocated Ge.

The data of Fig. 4 for dislocated Ge yield an even smaller value for the EHD diffusion constant,

$$D \lesssim 10^{-9} \text{ cm}^2\text{sec}^{-1} \qquad (3)$$

at 2.1K due to the smaller initial width of the EHD cloud. This measurement, 6 to 11 orders of magnitude smaller than previous experimental results,[1,2,3,6] is made possible by the lack of significant perturbing forces, and the sensitivity of this method. The extreme smallness of D from Eq. (3) implies that EHD must be pinned to crystal defects. Other than defects, the strongest damping mechanism for drop motion is thought to be phonon scattering.[1,11] Using the calculated[12] momentum relaxation time $\tau_p = 1.6$ nsec at 2.0K, we estimate that the diffusion coefficient due to phonon scattering is $D_p = \tau_p kT/M\upsilon \gtrsim 10^{-4}$ cm^{2}sec^{-1} with $M \simeq m_0$, for drops of total size υ less than 10^7 eh pairs.[13]

Because the measured valve of D from Eq. (3) is more than five orders of magnitude smaller than D_p, we rule out phonon scattering as the active damping mechanism.

Both crystals studied were high-quality ultrapure Ge.[8] Of the residual defects present in dislocated Ge, shallow impurity atoms are probably most important as pinning sites for EHD; the data of Figs. 3 and 4 make pinning on dislocations unlikely. We present a simple model for the diffusion of EHD in which individual drops move by hopping from one impurity atom to the next. Because the impurities in ultra-pure Ge are typically separated by several μm, a distance larger than the expected drop radius R, each hop consists of a thermally-activated escape from one impurity atom, followed by capture by another impurity atom after travelling a mean free path ℓ. Using the escape rate $(\bar{v}/R)\exp(-B/kT)$, the diffusion constant is

$$D_i = (1/3) \ell^2 (\bar{v}/R) \exp(-B/kT) \quad , \tag{4}$$

where $\ell = 1/\pi R^2 N_i$ with $N_i \sim 10^{11}$ cm^{-3} the total impurity concentration, $\bar{v} = (8 kT/\pi M \upsilon)^{1/2}$ is the drop thermal speed, and B is the binding energy of a drop on one impurity atom.[13] From the data of Figs. 3 and 4, we estimate the lower limit to the binding energy $B \gtrsim 5$ meV. This result is consistent with the calculated values[14] for shallow donors B = 5 to 7 meV. At higher T one might expect the diffusion constant from Eq. (4) to increase dramatically, but in fact D_i remains less than 10^{-9} cm^{2}sec^{-1} up to 4K for B = 5 meV, due to the strong size[13] dependence of Eq. (4).

Once sufficient external force is applied to EHD bound to defects, they break loose and travel through the crystal with the mobility corresponding to D_p above as observed by Alekseev et al.[6] and others.[1,5] We can roughly estimate the force of the phonon wind on EHD in our experiment by scaling the experimental results of Bagaev et al.[5] Assuming the entire excitation power $P \sim 2$ mWcm^{-2} from Figs. 3 and 4 is converted into phonons, and using[12] $\tau_p = 1.6$ nsec, and[7] $R_- \sim 0.2$ μm, we find $F_p \sim 3 \times 10^{-14}$ dyne for the force on an entire drop due to the phonon wind at 2.0K. This is a factor of $\sim 10^5$ smaller than the force $F_i \sim B/d \sim 6 \times 10^{-9}$ dyne necessary to remove a drop from an impurity atom, where $d \sim 20$ nm is the EHD surface thickness. The force on EHD under the same conditions due to the FE wind[4] is $F_{FE} \sim 3 \times 10^{-16}$ dyne, a factor $\sim 10^2$ smaller than F_p above. Thus we expect EHD to remain rigidly pinned to impurities at the excitation levels used in this experiment, in agreement with our data. At higher T the phonon wind force F_p increases dramatically due to increases[7,12,13] in the threshold excitation P_+ and drop radius R, the and its coupling with phonons; at 3.5K. F_p is comparable to F_i above, and sufficient to remove EHD from their pinning sites. This result is consistent with the experimental disappearance[7] of hysteresis effects at temperatures $T \gtrsim 3$K.
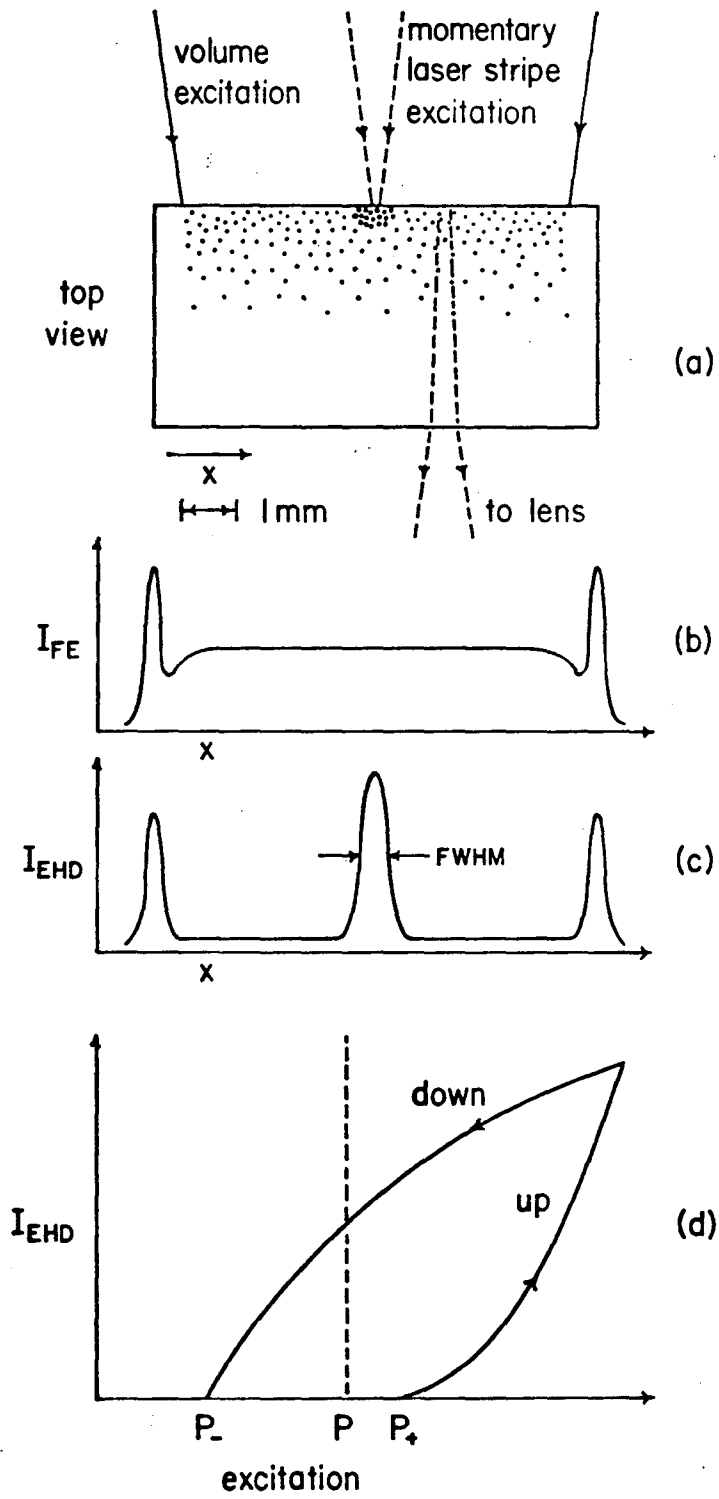
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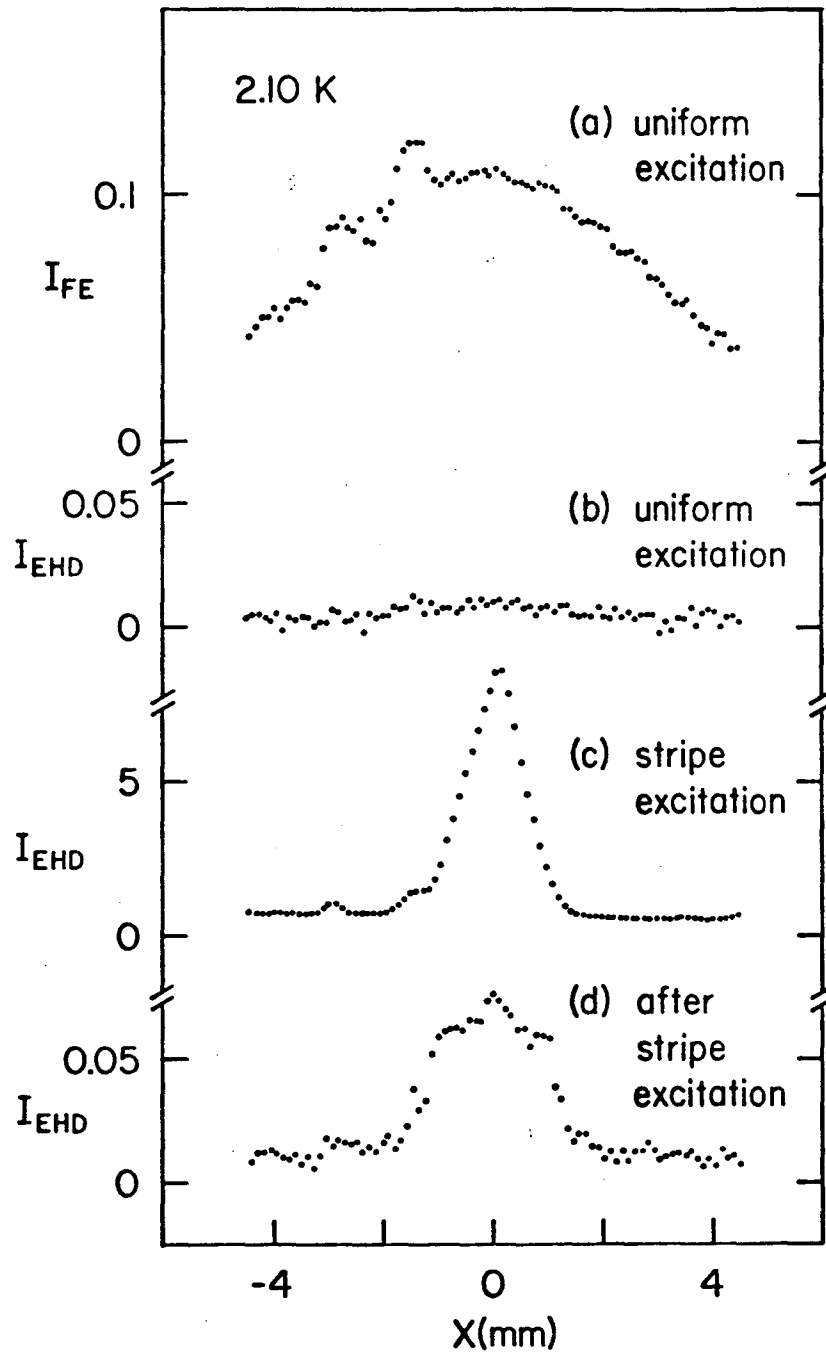
FIGURE CAPTIONS

Fig. 1. Experimental apparatus and technique: (a) excitation and
luminescence collection geometry showing Ge sample;
(b),(c) schematic scans of the free exciton (FE) and
electron-hole drop (EHD) luminescence intensities vs horizontal
position x; the peaks at the ends of each scan represent
scattering at the sample edges; (d) schematic hysteresis
curves for I_{EHD} vs excitation (see text).

Fig. 2. Scans of the luminescence intensities I_{FE} and I_{EHD} in mV
for sample 145M at 2.10K under various excitation conditions,
taken in sequence: (a),(b) uniform volume excitation monotonically
increased to the absorbed power $P \simeq 1.2$ mWcm^{-2}, no stripe
excitation; (c) during additional stripe excitation, absorbed
power P_{stripe} ~40 mWcm^{-1}; (d) after end of stripe excitation.

Fig. 3. Scans of the EHD luminescence intensity showing the EHD spatial
profile at various mean times following the end of stripe
excitation as indicated. Taken for sample 145M at 2.10K
with the same excitation sequence as for Fig. 2d.

Fig. 4. Scans of the EHD luminescence intensity as in Fig. 3 for
dislocated Ge, sample 437D, at 2.10K showing the EHD spatial
profile at various times following the end of stripe excitation:
uniform excitation absorbed power $P \simeq 2.4$ mWcm^{-2}, stripe
excitation absorbed power P_{stripe} ~70 mWcm^{-1}.

volume excitation

momentary laser stripe excitation

top view

(a)

x

⊢—⊣ 1mm

to lens

I_{FE}

(b)

x

I_{EHD}

FWHM

(c)

x

I_{EHD}

down

up

(d)

P_- P P_+

excitation

XBL 787-9550

Fig. 1

2.10 K

(a) uniform excitation

I_{FE}

(b) uniform excitation

I_{EHD}

(c) stripe excitation

I_{EHD}

(d) after stripe excitation

I_{EHD}

X(mm)

Fig. 2
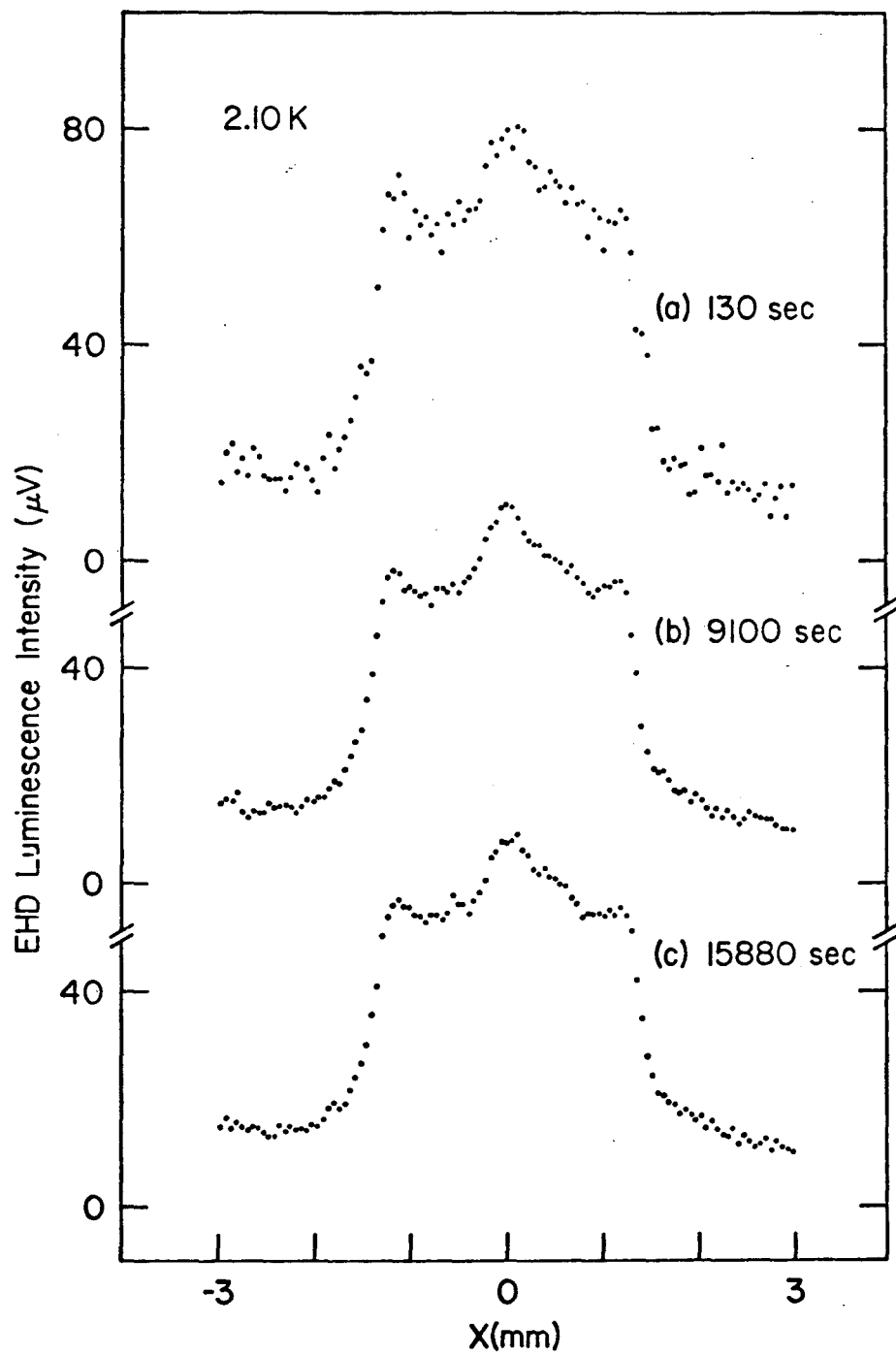
XBL 787-9549

Fig. 3

XBL 787-9547

Fig. 4