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POLARIZED PHOTOELECTRONS FROM OPTICALLY MAGNETIZED SEMICONDUCTORS*

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ABSTRACT

Electrons excited from the top of the spin-orbit-split valence band to the bottom of the conduction band by circularly polarized light are known to be spin polarized in materials such as GaSb and GaAs. By application of alternating layers of Cs and O_2 to GaAs, a negative electron affinity surface can be produced which permits electrons from the conduction band minimum to escape into vacuum, where we propose to measure their polarization by Mott scattering. Because of the high efficiency of negative electron affinity photoemitters, and the simplicity of electron production without applied magnetic fields, an electron source of 50% maximum polarization from GaAs appears attractive. We discuss criteria for a polarized electron source and compare the proposed GaAs source to the best existing source, photoemission from the ferro-magnetic semiconductor EuO. Some promising areas of investigation with a suitable source of polarized electrons are considered.

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I. INTRODUCTION

A source of electrons of high intensity and high degree of spin polarization would make possible a wide range of interesting experiments in many areas of physics. Ideally, one would like to have the intensity and electron optical properties of a good electron gun and in addition a 100% polarization of the electrons. After reviewing briefly some existing sources of polarized electrons (SPE) we discuss a proposal for a new, very promising source using optically magnetized GaAs with a negative electron affinity surface. The important characteristics of a SPE are discussed and the potential advantages of the GaAs source compared to existing sources are presented. Finally, we discuss some physical phenomena that could be investigated with a suitable source of polarized electrons.

II. SOURCES OF FREE POLARIZED ELECTRONS

Some physical phenomena giving rise to sources of free polarized electrons are listed in Table 1. These phenomena have been studied at least as much for their physical interest as for their potential as polarized electron sources. One of the most well known sources of polarized electrons is β-decay where the electrons are longitudinally polarized (i.e., along electron momentum) with a polarization v/c. However, even for very active sources, the intensities obtainable in an electron beam are very small.

When free electrons undergo elastic collisions with polarized atoms the polarization of the atoms can be transferred to the electrons through spin exchange.[1,2] Mott[3] was the first to suggest that electrons are polarized when scattered from unpolarized targets because the scattering cross section is spin dependent through the spin orbit interaction. At energies of 100 keV the scattering is due to the Coulomb field of the nucleus. Polarized electrons are also

obtained from scattering at energies in 1 keV range.[4] Here the de Broglie wavelength of the electrons becomes comparable with the effective extension of the atomic scattering potential and diffraction effects are observed giving rise to more complex variations of scattered intensity and polarization as a function of scattering angle. Resonant elastic scattering from Ne has also been used to produce polarized electrons.[5,6]

The scattering processes mentioned above do not yield sufficient intensity to be competitive SPE. More promising methods are based on the ejection of polarized electrons from atoms or solids. Polarized electrons can be ejected from atoms by collisional ionization, for instance of metastable polarized deuterium atom with a diamagnetic target gas, or as another example, in a He discharge involving metastable He atoms polarized by optical pumping.[8] A continuous current of a few μA and a polarization P $\sim$ 10% has been observed from the He discharge. If unpolarized alkali atoms are photoionized with circularly polarized light, polarized electrons are emitted (Fano effect).[9] Alternatively, polarized atoms can be photoionized with unpolarized light. This latter method has been developed at Yale University over the past 10 years and recently, from photoionization of a polarized Li atomic beam, an intensity of 2×10^8 electrons/1.5 μsec pulse with electron optical characteristics appropriate for injection in the Stanford Linear Accelerator Center (SLAC) electron accelerator was achieved.[10]

Emission of oriented electrons from solids has to date included field emission and photoemission as SPE. Field emission is attractive electron optically because the electrons emerge from the tip giving very nearly a point source. Field emission from EuS[11] coated W tips have produced electrons with P=80 to 90% but some development work remains before this can be treated as a reliable

source. In Zurich we have measured the spin polarization of electrons photoemitted from a number of solids, Fe, Co, Ni, EuO, EuS, and others, in order to study the fundamental nature of magnetism in these materials.[12] The highest polarization measured was for EuO, $P \simeq 80\%$, in an applied magnetic field of 30 kG as shown in Fig. 1.[13] EuO is potentially a very good SPE. The yield for photon energies 4 eV $\leq \hbar\omega \leq$ 6 eV is 3×10^{-3} electrons per photon which gives the photocurrent when the number of photons/sec at these energies incident on the sample is known. In a recent crash experiment to determine the suitability of EuO as a source for SLAC,[14] we measured 4×10^{8} electrons/1.5 μsec pulse which is already competitive with photoionization of Li. This is by no means an upper limit, as our apparatus is not designed as an electron source; a factor of 100 more could be obtained with an apparatus designed specifically as an SPE. However, we see that some experiments with polarized electrons can now be contemplated with a source like EuO. We turn now to our main subject, photoemission of polarized electrons from GaAs.

III. OPTICAL MAGNETIZATION OF GaAs

The optical orientation of gas atoms (optical pumping) has been extensively used to investigate atomic processes.[15] The interband absorption of circularly polarized light in a semiconductor with a spin orbit splitoff valence band can lead to a net spin orientation of nonequilibrium electrons in the conduction band. This was first observed in Si by Lampel[16] who used the dynamic nuclear polarization to monitor the electron polarization. The now more common method of measuring the polarization of the photoluminescence was first used by Parsons[17] on GaSb. The proposed SPE is based on a series of Russian works at the Ioffe Physico-Technical Institute which have investigated GaAs and ternary GaAs compounds.[18]

The origin of the spin polarization of the electrons in the conduction band is seen in Fig. 2. Semiconductors with a diamond or zincblende structure have a band structure at the centre of the Brillouin zone (Γ) as shown in the left side of the figure. The band gap energy E_g is 1.52 eV and the spin-orbit splitting is 0.33 eV for GaAs.[19] The momentum of the circularly polarized light $M=\pm 1$ is transferred to the electron hole system. The selection rules at k=0 are the same as for atomic transitions between states with $m_J=3/2$ and $m_J=1/2$. Considering the transitions for circularly polarized light with M=1, solid lines in Fig. 2, the probabilities for transitions from the top of the valence band (circled numbers in Fig. 2) are such that three times as many electrons are excited to states with spin opposed to the photon angular momentum as to states with spin parallel.[20] This gives rise to a polarization

$$P = \frac{n\uparrow - n\downarrow}{n\uparrow + n\downarrow} = \frac{3-1}{3+1} = 50\% \quad .$$

Due to the large number of holes in the p type materials used, there is negligible polarization of the valence band. The degree of polarization of the electrons in the conduction band depends on the relative magnitudes of the electron spin relaxation time τ_s and the electron lifetime τ_e. The measured polarization P is related to the theoretical polarization $P_{max}=50\%$ by

$$P/P_{max} = \tau_s/(\tau_e + \tau_s) \quad . \qquad (1)$$

Measurements of the polarization at 4.2°K from GaAs as a function of photon energy is shown in Fig. 3 from Ref. 21 for two different dopings. The decrease in measured polarization relative to the theoretical value (dashed line in Fig. 3) is given by Eq. (1). Electrons excited to energies above the conduction band minimum thermalize in about 10^{-12} sec. The increased scattering in heavily doped material retards spin relaxation during thermalization.[22] This is analogous

to dynamic narrowing of magnetic resonance lines. Electrons excited from the split off valence band contribute the opposite polarization (Fig. 2). Thus in the lightly doped material where the spin of hot electrons from Γ_8 relax rapidly, for $\hbar\omega > E_g + \Delta$, a negative polarization is observed due to the contribution from Γ_7 electrons. Note that in the heavily doped samples, a high polarization is maintained for excitation with photon energies up to 0.3 eV above threshold.

It is possible to determine the relaxation times τ_e and τ_s separately by making an additional measurement in a magnetic field transverse to the light polarization. This compares the total relaxation rate $\tau_0^{-1} = \tau_e^{-1} + \tau_s^{-1}$ with the Larmor frequency ($\omega = g\mu_B H/\hbar$) of precession in the field. The field dependence of the polarization is described by a Lorentzian

$$P(H) \propto 1/\left(1 + \omega^2\tau_0^2\right) \tag{2}$$

from which one obtains τ_0 assuming g is known. Typical results are shown in Fig. 4 from Ref. 23. For GaAs at 4.2°K, $\tau_s = 8 \pm 1.5 \times 10^{-10}$ sec and $\tau_e = 2.4 \pm 0.3 \times 10^{-10}$ sec.[23] The temperature dependence of τ_s has been investigated and is characteristic of spin lattice relaxation by acoustic phonons above 77°K. From Fig. 4 it is apparent that in $Ga_{0.7}Al_{0.3}As$ another spin relaxation mechanism is present at low temperature. This has been identified as spin relaxation through hyperfine interaction with the nucleus since τ_s increases in a longitudinal field which decouples the electron and nuclear spin.[24]

From the point of view of a SPE, it is sufficient to know that a significant polarization is observed, and reassuring that the relaxation times and depolarization mechanisms have been investigated. We see that a reasonable polarization may be attainable at liquid nitrogen temperature which would be more convenient than working at liquid helium temperature.

IV. NEGATIVE ELECTRON AFFINITY

Naturally, in order for the optically polarized electrons in GaAs to be suitable as a source, we must be able to get the electrons out into the vacuum. That requires exciting the electrons to states which are above the vacuum level which for a freshly cleaved (110) GaAs surface lies Ea = 3.2 eV above the conduction band minimum in the bulk (Ea is the electron affinity, see Fig. 5). The beauty of p-type GaAs is that by proper activation of the surface it is possible to lower the vacuum level to achieve a negative electron affinity.[25,26] Figure 5 shows the bands and vacuum level for GaAs for a clean surface, a surface with a monolayer of cesium, and a surface activated with cesium and oxygen. With cesium alone it is possible to lower E such that $E_a = O$. With Cs_2O the $E_a < O$ and the electron escape is limited by the interfacial barrier between the GaAs and the Cs_2O.[27] Photoexcited electrons diffuse rapidly to the conduction band minimum and then diffuse to the surface where they may escape. The photoyield is limited by the diffusion length L which is of the order of 1μ ($L_\Gamma = 1.2\mu$ at $T = 300^oK$, $\rho = 3\times10^{19}/cm^3$)[26] instead of by the hot electron scattering length which is of the order of 100Å. The electron does have to pass through the bandbending region where it is a hot electron and can scatter from optical phonons. Heavily doped material is used to obtain a narrow bandbending region.

For Cs on (110) GaAs, the valence band is pinned at the surface $V_{BB} = 0.6$ eV below its position in the bulk giving rise to a bandbending over a length of 80 Å for $\rho = 1\times10^{19}/cm^3$; in GaAs + Cs_2O where $V_{BB} = 0.23$ eV the corresponding bandbending is over 50 Å.[28]

The treatment of GaAs to obtain a negative electron affinity has been extensively studied because of its usefulness as a photocathode. Figure 6 shows, for example, the escape probability as a function of Cs and O_2 treatment.[29] For

p-GaAs ($1\times10^{19}/cm^3$) a maximum is observed at about 5 Cs and O_2 treatments corresponding to approximately 1 Langmuir O_2 exposure.

The yields of variously activated samples of GaAs[26,30] are compared in Fig. 7 to the yield from GaAs with a clean cleaved (110) surface[26] and from EuO with a (100) surface.[31] For our measurements we are interested in electrons excited at photon energies $E_g < \hbar\omega < E_g + 0.3$ eV. The best yield at threshold can be obtained from GaAs + Cs_2O although GaAs treated with CsF and Cs is also quite good.[30] The yield from the highly polarized 4f states of EuO, 4 eV $\leq \hbar\omega \leq$ 6 eV, is seen to be two orders of magnitude below that of GaAs. In the case of GaAs, a yield of 0.2 eV electrons/photon at $\hbar\omega \sim E_g$, means that with 1 mW of light onto the sample a dc photocurrent of 100 μA would be generated.

If a photoexcited electron escapes from the solid it does so within a time τ_e; since we know that $\tau_e < \tau_s$, the electron should be polarized as has been measured in luminescence measurements. The big question is whether the electron is depolarized as it crosses the Cs_2O layer. We plan to test this experimentally by measuring the spin of the photoemitted electrons using Mott scattering. A depolarization due to scattering in the Cs_2O layer can be investigated by varying the thickness of the Cs_2O layer. Initially, clean GaAs surfaces will be obtained by cleaving GaAs single crystals. We envision that a practical source would use epitaxially grown films of GaAs, GaAlAs, or GaAsP which would be cleaned by heating to just below the decomposition temperature before the activation process was started. The yield of Cs_2O itself at these photon energies is $\sim4\times10^{-3}$ and thus negligible compared to the activated GaAs.[27]

V. CHARACTERISTICS OF A POLARIZED ELECTRON SOURCE

In this section we consider the characteristics of a SPE and compare the expected values for GaAs + Cs_2O as a potential source to EuO, the best existing

SPE from a magnetic material. A list of characteristics to consider is given in Table 2. The polarization of GaAs (we refer to GaAs + Cs_2O as the GaAs source) would be ~40% compared to ~80% for EuO. In contrast to EuO where a large magnetic field must be reversed to reverse the polarization, for GaAs the polarization is simply reversed by rotating the polarizer. In fact it is possible to rotate the light polarizer at some convenient frequency, say 70 Hz, and use synchronous detection techniques to measure very small changes as a function of change in polarization. The intensity to be expected depends very much on the light source whether pulsed or dc. What can be said is that the yield of GaAs is 100 times that of EuO. The spread in energy of the GaAs source must be <0.3 eV and in the EuO source can be as large as 2 eV; this can give additional intensity from the EuO at the expense of a well defined beam energy. In experiments where the statistical accuracy of the measurements is the crucial factor, a figure of merit for a source is the product P^2I; in these circumstances what GaAs lacks in P can easily be made up by the higher intensity. In the case of pulsed measurements the time structure is important and determined primarily by the lamp. Since a significant polarization is observed in GaAs at $77^{\circ}K$ and activated GaAs cathodes are stable at room temperature, GaAs does not appear to be at a disadvantage relative to EuO with a Curie temperature of $69^{\circ}K$.

We come now to the electron optical characteristics of the source as given by the emittance (aperture angle × radius of beam) at a given electron energy. This is a property of the electron beam that cannot be changed and is the result of the law of Helmholtz-Lagrange[32]

$$d\Omega_1 dA_1 E_1 = d\Omega_2 dA_2 E_2 \qquad (3)$$

where $d\Omega$ is the differential solid angle, dA the differential area, and E the beam energy all at beam cross section 1 and 2 (assuming current is conserved along the beam path). The emittance is given by

$$\epsilon = r_0(E_0/E)^{1/2} \tag{4}$$

where r_0 is the distance of the electron point of origin from the beam axis, E_0 is the initial energy and E the final energy. In the case that a magnetic field is present at the point of electron origin another contribution is present, which arises physically from the fact that the z component of the canonical angular momentum,

$$\vec{L} = \vec{r}_0 \times (m\vec{v} - e\vec{A})$$

where m is the mass, e the charge, v the electron velocity and $\vec{A}$ the vector potential, is conserved. This leads to a skewing of the electron trajectories when they are in a region of zero magnetic field giving an effective emittance,[10]

$$\epsilon \simeq r_0(E_0/E)^{1/2} + 1/2(e/m)\, r_0^2 B_0/v \tag{5}$$

where B_0 is the magnetic field at the electron point of origin. Since a high field is present when EuO is in magnetic saturation, it is more suited for experiments at high energies in order to reduce the contribution of the second term in Eq. (5), when a given emittance is required. This term can also be decreased by reducing r_0 which can best be done by focussing a laser beam to a small area on the sample, the main limitation being the local heating of the sample. By frequency doubling the 5145 Å line of an argon ion laser high intensity can be achieved at 4.8 eV. EuO is also suitable as a SPE when the experiment itself is performed in a region of high magnetic field.

In the case of the GaAs source, no magnetic field is present and the emittance is given by Eq. (4). GaAs is especially suitable for low energy electron beam experiments in field free regions. The emittance can be kept small even at low energies by minimizing r_0 with a well focussed light spot.

VI. EXPERIMENTS WITH POLARIZED ELECTRONS

We now turn to a brief discussion of a few of the possible areas of experimental study using polarized electrons. Much of the impetus and funding for the development of SPE has come from high energy physics which we won't go into here.

Suitable SPE would make feasible such experiments, for example, as the measurement of the spin structure of the scattering amplitude for deep inelastic scattering of polarized electrons by polarized protons or the study of possible contributions from weak parity-violating interactions in the scattering of polarized electrons by unpolarized protons in a liquid hydrogen target.[33]

There have been a number of investigations of spin dependent scattering of electrons with energies ranging from 100 eV to 100 keV, from atoms, molecules and solids.[2,4] At these energies the spin dependence of the scattering arises from the spin-orbit interaction (Mott scattering). Qualitatively, the scattering potential is made up of the spin-orbit potential and the Coulomb potential, either of the nucleus at higher energies or of the screened nucleus at lower energies. Up until now, all experiments have been double scattering experiments: an unpolarized electron beam is scattered from the target and the polarization of the scattered electrons is measured in a second scattering experiment. In the case of elastic scattering from unpolarized targets it is entirely equivalent if one has an unpolarized incident beam and measures the polarization of the

scattered electrons, or if one has a polarized incident beam and measures the asymmetry in the intensity of the scattered electrons. Thus, a source of polarized electrons eliminates the second scattering. The importance of this can be grasped when it is realized that the efficiency of a Mott detector to measure the polarization is very low; the ratio of incident to measured electrons is about 10^{-4}. Experiments at higher resolution as well as new types of experiments can be considered when a suitable SPE is available.

For electron scattering at lower energies ($\leq$ 100 eV) from magnetic materials the exchange interaction makes an important contribution to the spin dependence in the scattering.[34] One envisages, for example, the scattering of a well focussed electron beam from a single domain of magnetic material. The exchange scattering depends on the number, distribution and magnitude of spins in a unit cell. Evidence for coherent exchange scattering was found in LEED studies of NiO above and below the Neel temperature T_N.[35] The magnetic cell of NiO has dimensions twice the chemical cell at $T < T_N$ which gives rise to half order diffraction beams which disappear at T_N. The intensity of the exchange scattered beams was 1 - 3% of the integer order beam. In crystals where the spin structure does not differ from that of the Coulomb potential, the exchange and Coulomb intensities interfere. However with a polarization-modulated incident electron beam, one could easily measure 1 - 3% intensity variations in the diffracted beams. Since the exchange interaction is very sensitive to the nature of the wave functions, polarized LEED measurements can be expected to yield interesting new information.

Polarized LEED measurements need not be restricted to magnetic materials where the exchange interaction is important. Recent calculations by Feder[36] for W show that even at low energies the spin orbit interaction gives rise to

significant spin-dependent scattering. The coherent nature of the scattering could also be exploited for a SPE as first suggested by Maison.[37] Alternatively, such scattering from a well characterized W single crystal surface can be used as a low energy polarization detector.

In summary, of the many phenomena which have been investigated as sources of polarized electrons, photoemission from EuO stands way ahead at present. The proposed GaAs source with a negative electron affinity surface, if there is no serious depolarization by the Cs_2O layer, would approach the goal of achieving the characteristics of a good electron gun that, in addition, delivers polarized electrons. Clearly the number of new experiments which would then be possible is enormous.

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Table 1. Sources of Free Polarized Electrons

- Beta Decay
- Electron Spin Exchange with Polarized Atoms
- Electron Scattering from Unpolarized Targets
  - High-Energy Mott Scattering
  - Low-Energy Mott Scattering
  - Resonant Elastic Scattering
- Ejection of Electrons from Atoms
  - Collisional Ionization of Polarized Metastable Atoms
  - Photoionization of Unpolarized Alkali Atoms by Polarized Light
  - Photoionization of Polarized Alkali Atoms
- Emission from Solids
  - Field Emission from EuS Coated W Tip
  - Photoemission from Magnetized Solids: Fe, Co, Ni, EuO, EuS, etc.
  - Photoemission from Nonmagnetic Semiconductor, eg., GaAs, by Polarized Light

Table 2. Characteristics of a Spin Polarized Electron Source

- Polarization
  - Degree
  - Reversal
- Intensity
  - DC
  - Pulse
- Energy
  - Mean
  - Spread
- Time Structure
  - Pulse Length
  - Repetition Rate
- Emittance

FIGURE CAPTIONS

1. Spinpolarization of electrons photoemitted from nonintentionally doped EuO and EuO + 2% La are shown as a function of magnetic field. The vertical error bars represent $\pm\sigma$ in the counting statistics. The polarization of Ni is shown for comparison (from Ref. 13).

2. Bandstructure E vs k at k=0 for diamond and zincblende semiconductors showing the spin-orbit split-off valence band. The transitions for circularly polarized photons with angular momentum $M = \pm 1$ ($\sigma_{\pm}$) are shown for σ^{+}(——) and σ^{-}(- - - -), where the relative transition probabilities are indicated by the circled numbers. For photon energies $E_g < \hbar\omega < E_g + \Delta$, the theoretical optically-induced spin polarization in the conduction band minima is 50%.

3. The theoretical polarization in GaAs as a function of photon energy (dashed line) is compared to experimental results from p-type GaAs crystals with two different dopings (from Ref. 21).

4. The magnetic field dependence (transverse to light beam) for GaAs and $Ga_{0.7}Al_{0.3}As$ both at $4.2^{o}K$ and $77^{o}K$ (from Ref. 23).

5. Energy level diagram showing the surface of a freshly cleaved GaAs crystal, GaAs with a monolayer of Cs which gives electron affinity $E_g \sim O$, and GaAs - Cs_2O which gives a negative electron affinity.

6. The escape probability for electrons at the conduction band minimum in the center of the Brillouin zone Γ as a function of the number of Cs and O_2 treatments. O_2 exposure and Cs_2O thickness are also indicated (from Ref. 29).

7. The yield of clean and variously activated GaAs surfaces (Refs. 26 and 30) and the yield of EuO (Ref. 31) are compared. Note that GaAs is about two orders of magnitude higher than EuO.

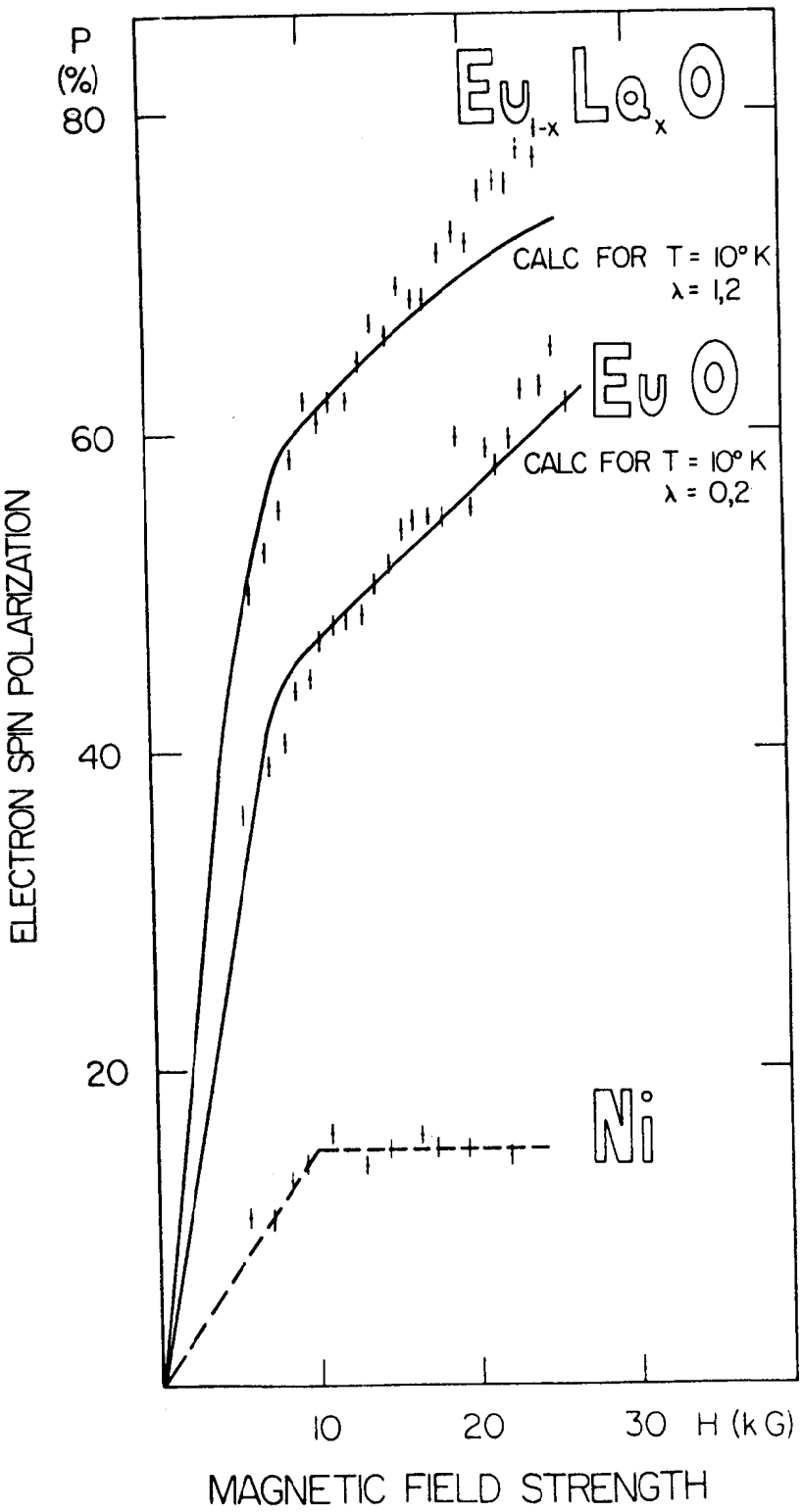
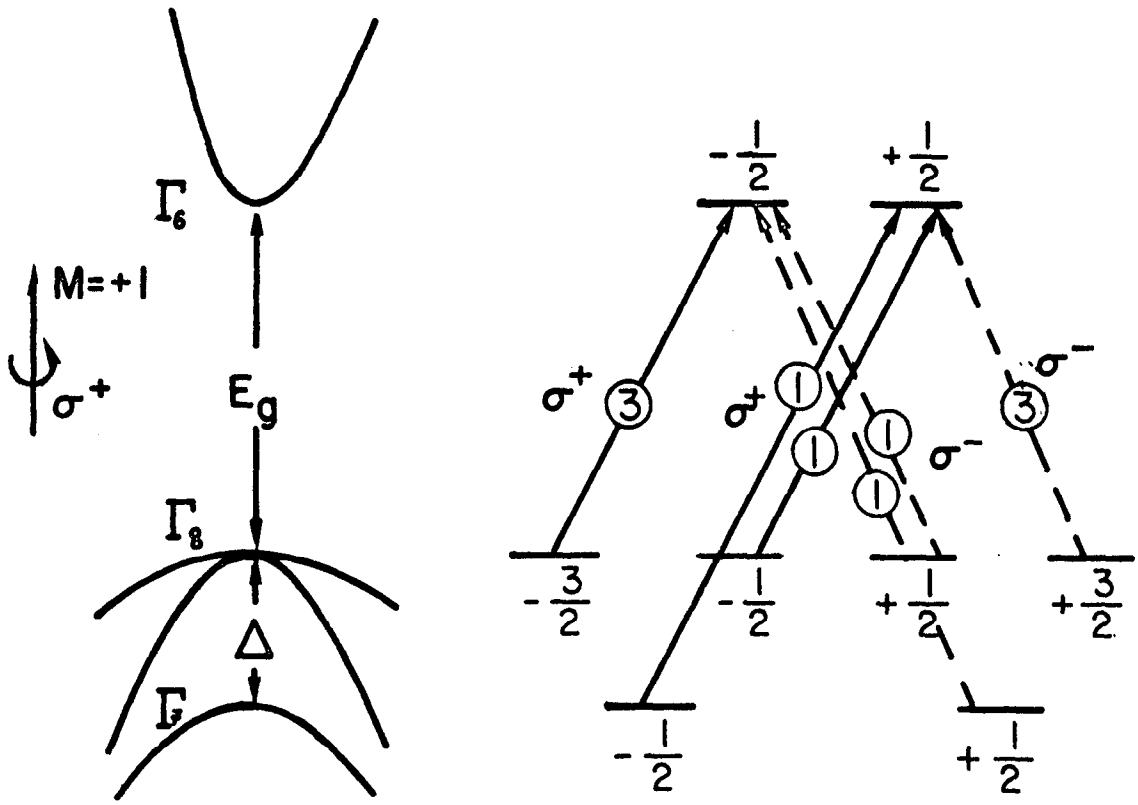

P
(%)
80
60
40
20
ELECTRON SPIN POLARIZATION
Eu1-x Lax O
CALC FOR T = 10° K
λ = 1,2
Eu O
CALC FOR T = 10° K
λ = 0,2
Ni
10
20
30 H (kG)
MAGNETIC FIELD STRENGTH

Fig. 1



Energy levels and transition probabilities in GaAs

Fig. 2

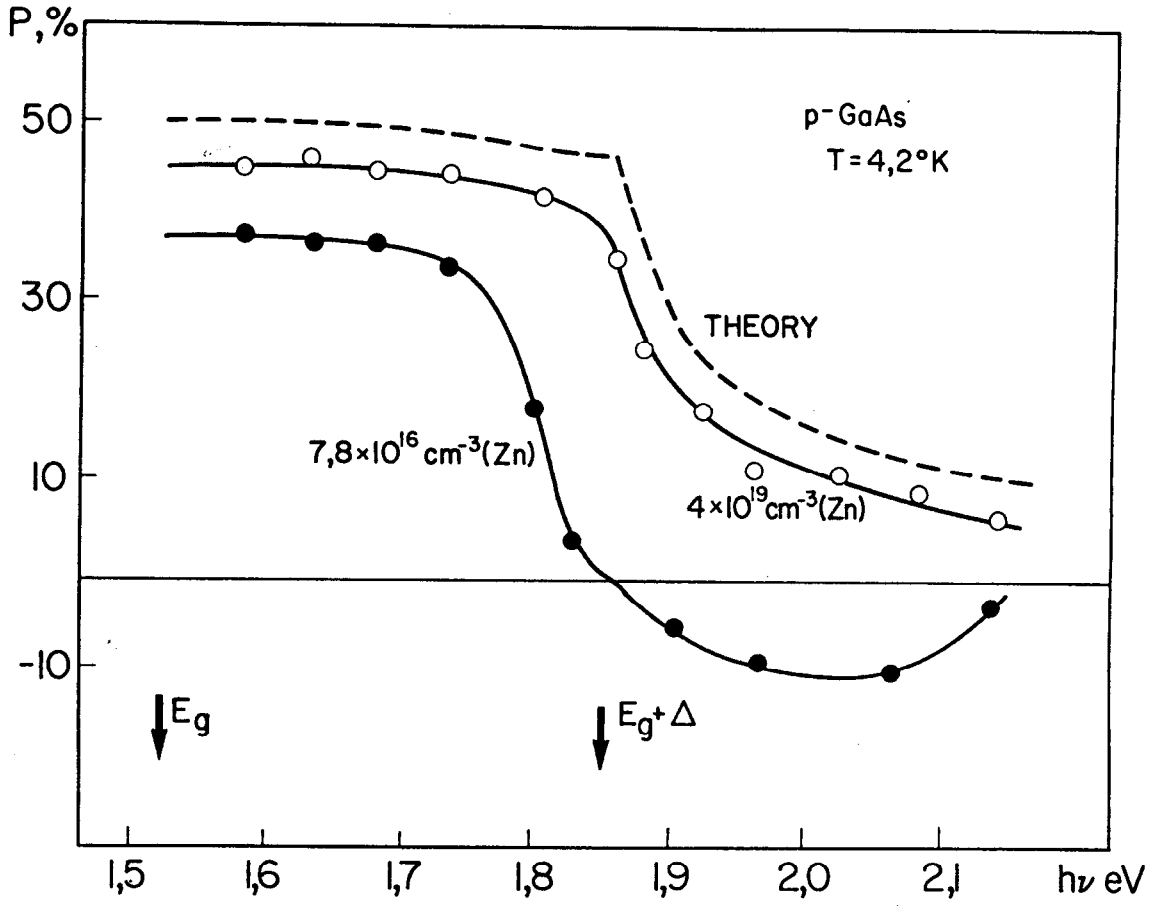

P,%
50
30
10
-10
p-GaAs
T=4,2°K
THEORY
7,8×10^16 cm^-3(Zn)
4×10^19 cm^-3(Zn)
Eg
Eg+Δ
1,5
1,6
1,7
1,8
1,9
2,0
2,1
hν eV

Fig. 3

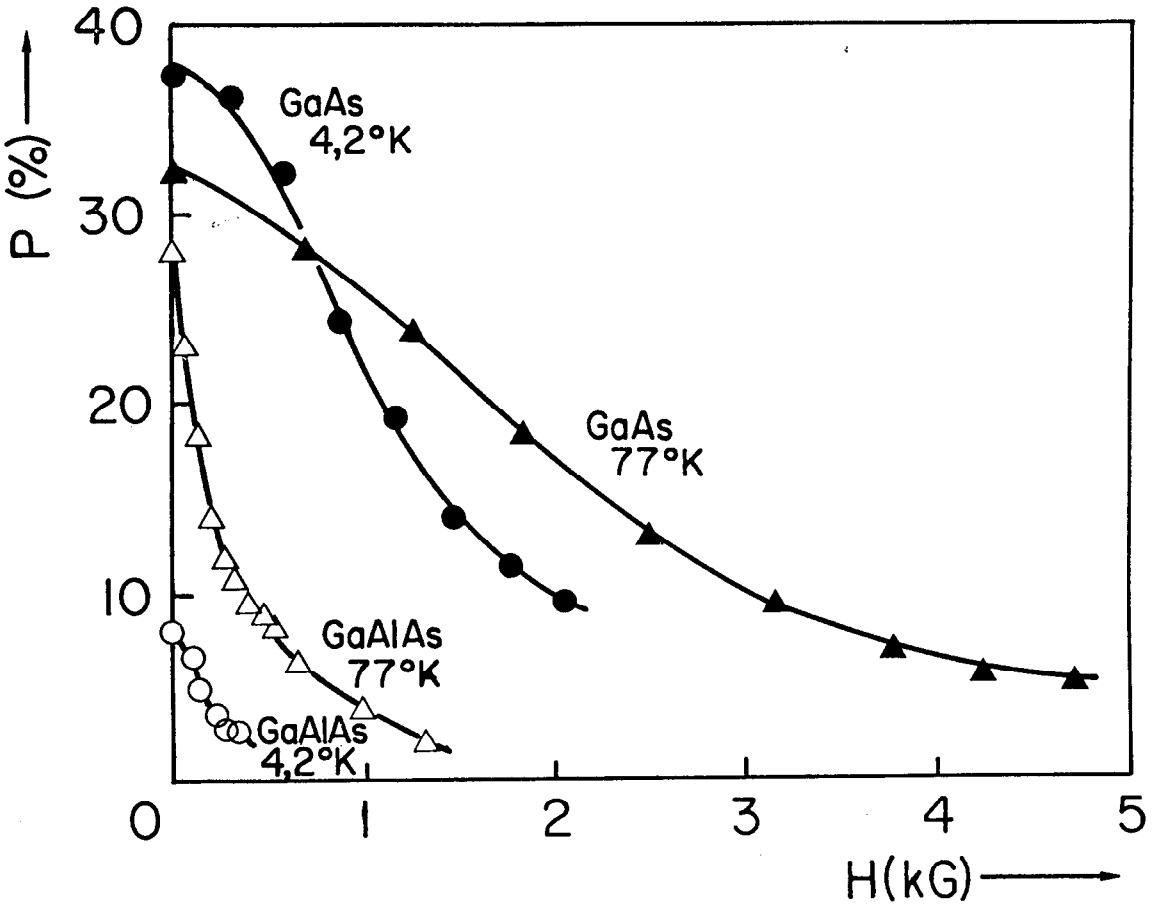
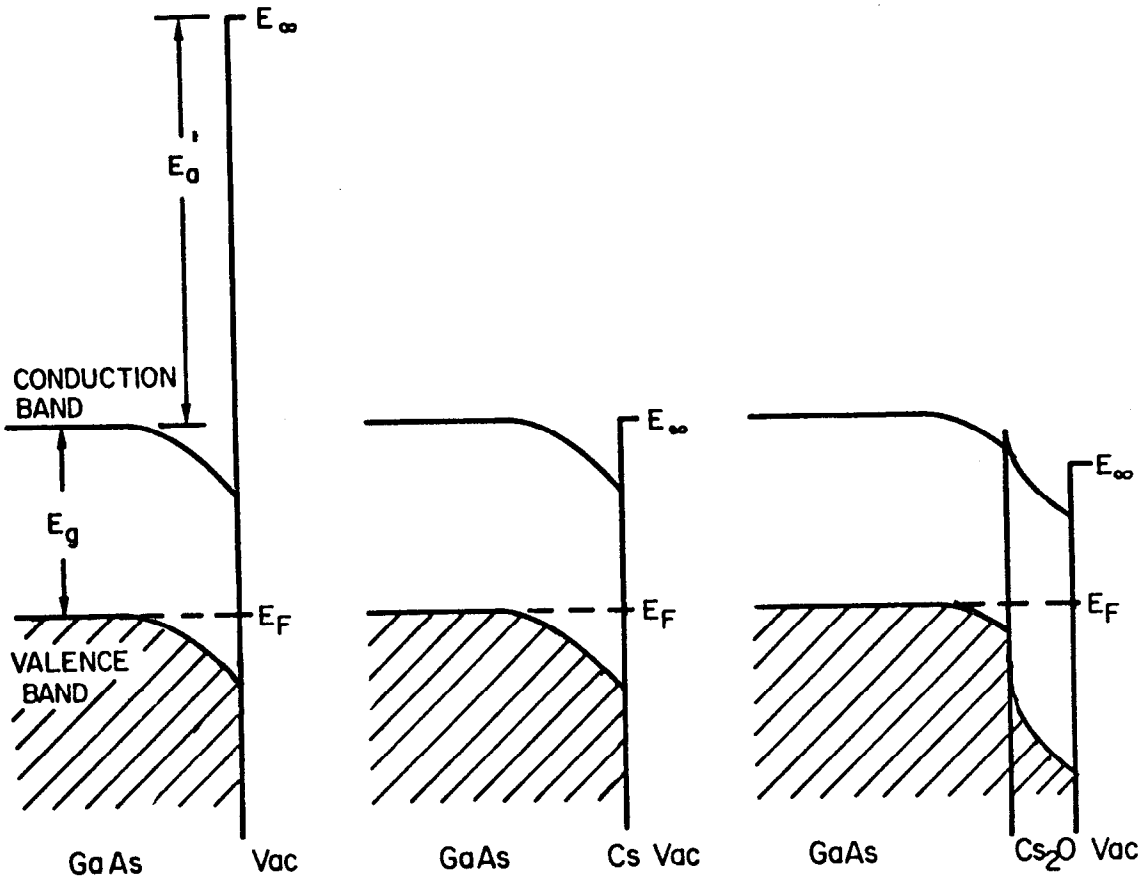

40
30
20
10
0
1
2
3
4
5
P (%)
H(kG)
GaAs
4,2°K
GaAs
77°K
GaAlAs
77°K
GaAlAs
4,2°K

Fig. 4



Energy band diagram of GaAs with a clean surface and treated to produce a negative electron affinity

Fig. 5

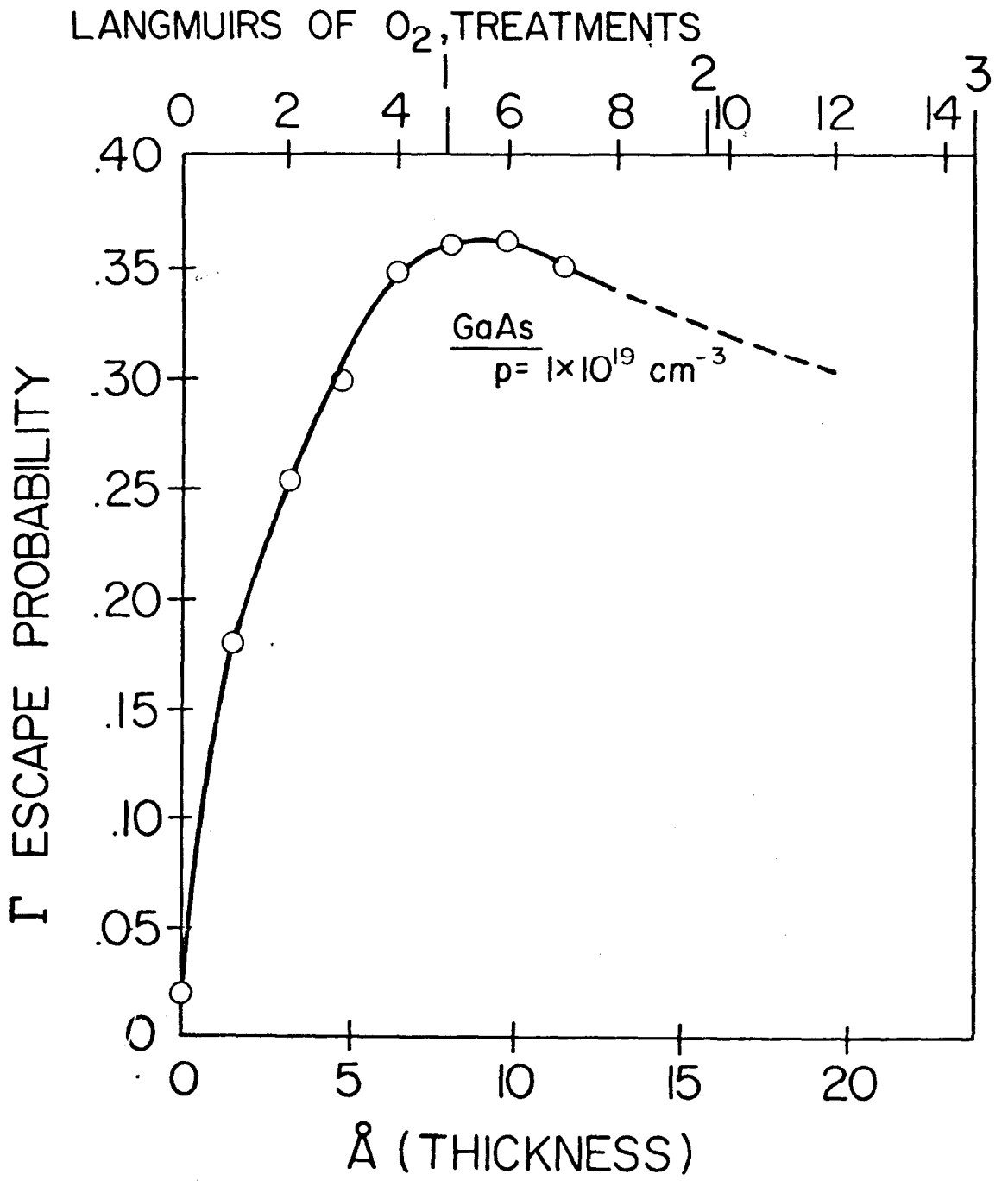

LANGMUIRS OF O_2, TREATMENTS
1
2
3
0
2
4
6
8
10
12
14
.40
.35
.30
.25
.20
.15
.10
.05
0
Γ ESCAPE PROBABILITY
GaAs
p= 1×10^19 cm^-3
0
5
10
15
20
Å (THICKNESS)

Fig. 6

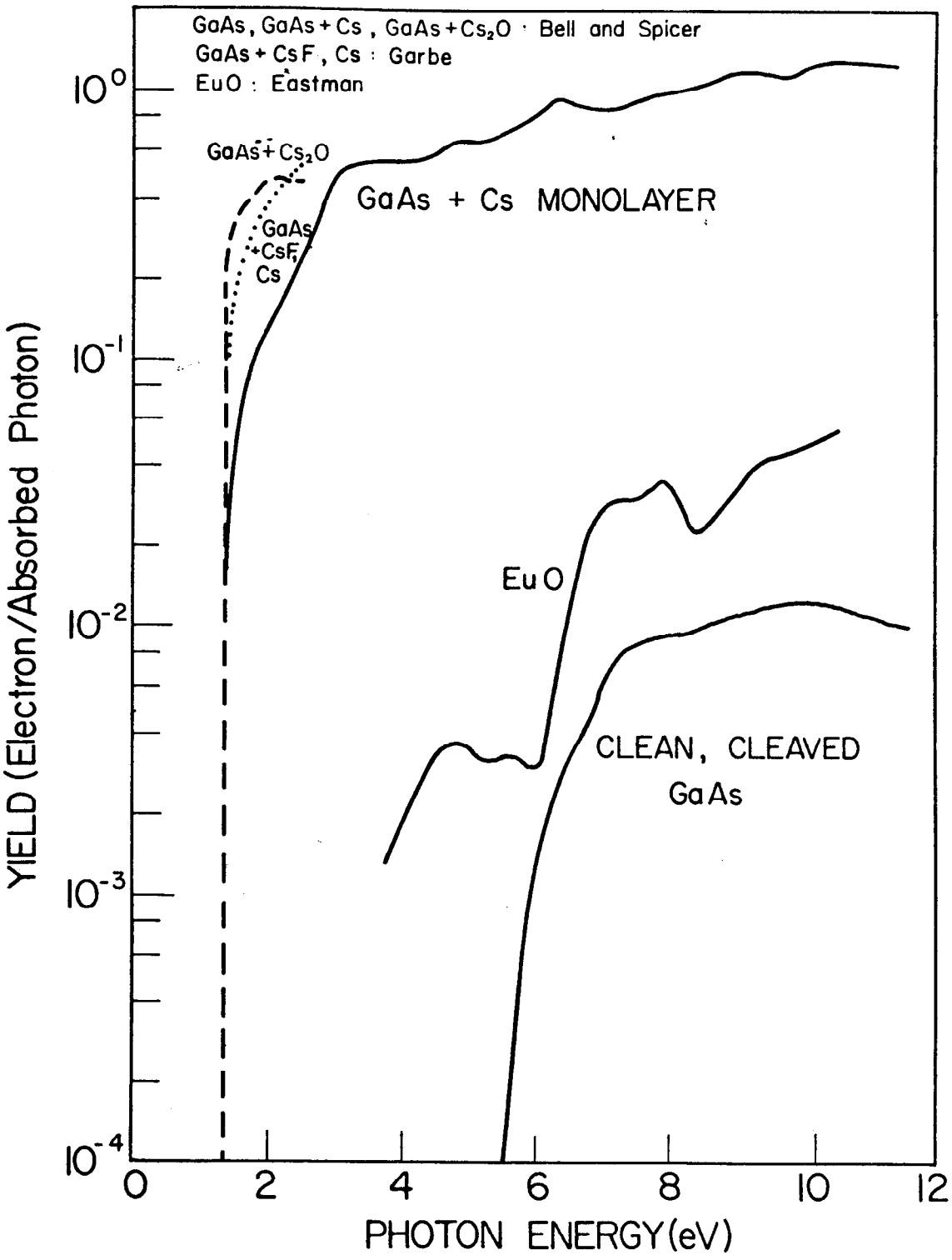

GaAs, GaAs + Cs, GaAs + Cs2O · Bell and Spicer
GaAs + CsF, Cs : Garbe
EuO : Eastman
GaAs + Cs2O
GaAs + CsF, Cs
GaAs + Cs MONOLAYER
EuO
CLEAN, CLEAVED GaAs
YIELD (Electron/Absorbed Photon)
PHOTON ENERGY (eV)
10^0
10^-1
10^-2
10^-3
10^-4
0
2
4
6
8
10
12

Fig. 7