Spontaneous Spin Polarization of Photoelectrons from GaAs

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We observe a spontaneous electron-spin polarization in GaAs(110) in photoemission experiments using excitation with linearly polarized light. This effect is a consequence of the noncentrosymmetric crystal structure of GaAs and suggests that the polarization of each spin-split conduction band can be almost as high as ± 100%. A net nonzero polarization after summation over spin states arises from (a) differences in spin-up and -down conduction-band hybridization with valence p states, and (b) surface-transmission effects.

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We report on a new effect in photoemission which creates spin-polarized electrons without making use of magnetically ordered electron spins (as in photoemission from magnetic materials) or of optical spin orientation by excitation with circularly polarized light. This new effect has its origin in the well-known lack of space-inversion symmetry in III-V compounds. There have been previous reports of spin-polarized electron emission, by excitation with linearly polarized light, from Xe atoms[1] or from a W(001) surface.[2] In those cases, however, the observed effect arises either from the angular correlation between electron polarization and the direction of the exciting radiation, or from spin-dependent diffraction in off-normal emission, respectively.

Quite generally, as a result of time-reversal symmetry, the energy eigenvalues of electronic states have to fulfill

$$E(\mathbf{k},\uparrow)=E(-\mathbf{k},\downarrow),$$
$$E(\mathbf{k},\downarrow)=E(-\mathbf{k},\uparrow), \tag{1}$$

where $\mathbf{k}$ is the electron wave vector and $\uparrow$, $\downarrow$ denote the spin states. In crystals like germanium, which additionally have space-inversion symmetry, $E(\mathbf{k},\uparrow)=E(-\mathbf{k},\uparrow)$ must hold and therefore with Eq. (1) $E(\mathbf{k},\uparrow)=E(\mathbf{k},\downarrow)$; i.e., all states are at least doubly degenerate. However, GaAs crystallizes in the zincblende structure, which does not have this inversion symmetry. Therefore a spin splitting of the energy bands arises. The classical works of Dresselhaus,[3] Parmenter,[3] and Kane[4] demonstrate how this splitting occurs for all $\mathbf{k}$ except for the $\langle 100\rangle$ and $\langle 110\rangle$ lines. We demonstrate in the present work that as a result of this effect it is possible to observe emission of spin-polarized electrons from GaAs(110) by photoexcitation with *unpolarized* radiation. Further, we shall compare the measured polarization to results obtained from first-principles calculations. This serves the purpose of estimating the importance of the intrinsic band-structure (bulk) polarization as compared to surface-transmission–induced effects.

The spin splitting of the lowest conduction band (∇_6 state at $k=0$) in GaAs for $\mathbf{k}$ along [110] is considered. As stated above, the splitting must, as a result of symmetry requirements, be zero at two points, Γ and $X=(1,1,0)2\pi/a$, on this line. Approximately one-third of the way along Γ-X, it attains a maximum. The dispersion is illustrated in Fig. 1,

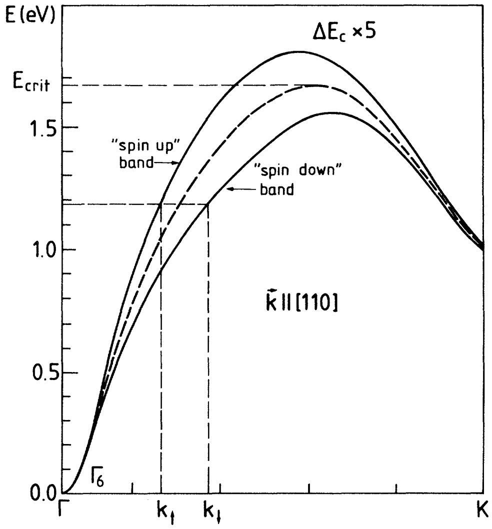

FIG. 1. Lowest conduction band (Γ_6 state at zone center) along Γ-K calculated by means of the relativistic linear-muffin-tin-orbital method. The spin-splitting ΔE_c has been exaggerated by a factor of 5. The dashed line shows the band without spin splitting.

where, however, only the part Γ-K is included.

In the limit of small k, perturbation theory shows that the conduction states can be fully polarized in a direction $\mathbf{\Omega}/|\mathbf{\Omega}|$ perpendicular to $\mathbf{k}$. To see this, consider the perturbing Hamiltonian of D'yakonov and Perel',[5]

$$H' = (\hbar/2)\boldsymbol{\sigma}\cdot\mathbf{\Omega}, \tag{2}$$

where $\boldsymbol{\sigma}$ is the Pauli spin operator, and $\mathbf{\Omega} = \gamma(k_x(k_y^2-k_z^2), k_y(k_z^2-k_x^2), k_z(k_x^2-k_y^2))$, with γ dependent[5] on band-structure parameters at Γ, but k independent. The degenerate states of the Γ_6 band are $iS|\uparrow\rangle'$ and $iS|\downarrow\rangle'$. For $\mathbf{k}=k(\sin\alpha\cos\beta, \sin\alpha\sin\beta, \cos\alpha)$ the spin part of the wave function is[4]

$$\begin{aligned} |\uparrow\rangle' &= e^{-i\beta/2}\left\{\cos\frac{\alpha}{2}|\uparrow\rangle + e^{i\beta}\sin\frac{\alpha}{2}|\downarrow\rangle\right\}, \\ |\downarrow\rangle' &= e^{-i\beta/2}\left\{-\sin\frac{\alpha}{2}|\uparrow\rangle + e^{i\beta}\cos\frac{\alpha}{2}|\downarrow\rangle\right\}, \end{aligned} \tag{3}$$

where $|\uparrow\rangle, |\downarrow\rangle$ are eigenstates of

$$\sigma_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}.$$

Here the quantization axis is arbitrarily taken along z. With the perturbation turned on the spin wave function becomes

$$|\psi\rangle = a|\uparrow\rangle' + b|\downarrow\rangle' = a|\psi_1\rangle + b|\psi_2\rangle. \tag{4}$$

In the case $\mathbf{k}\parallel[110]$ one gets, using Eq. (2),

$$\boldsymbol{\sigma}\cdot\mathbf{\Omega} = (\gamma/\sqrt{2}\hbar)k^3 S_\Omega, \tag{5}$$

where $S_\Omega = \sigma_x - \sigma_y$. The matrix elements of this operator are $\langle\psi_i|S_\Omega|\psi_i\rangle = 0$ and $\langle\psi_1|S_\Omega|\psi_2\rangle = i\sqrt{2}$, and thus the 2×2 secular determinant yields the spin splitting $\Delta E_c = \gamma k^3$ (valid for small k, see also Fig. 1). Furthermore $a = \pm ib$ which gives the wave functions

$$\begin{aligned} |\psi_\uparrow\rangle &= 1/\sqrt{2}(i|\psi_1\rangle + |\psi_2\rangle), \\ |\psi_\downarrow\rangle &= 1/\sqrt{2}(|\psi_1\rangle + i|\psi_2\rangle), \end{aligned}$$

which are pure spin-"up" and -"down" states with respect to the $[1\bar{1}0]$ direction [see Eqs. (3) and (4)]. The operator for spin quantization along $\mathbf{\Omega}$ is $\sigma_\Omega = \boldsymbol{\sigma}\cdot\mathbf{\Omega}/|\mathbf{\Omega}|$, and the polarization $\mathbf{P}_\Omega = \langle\psi_i|\sigma_\Omega|\psi_i\rangle/\langle\psi_i|\psi_i\rangle = \pm 100\%$ for $i = \uparrow, \downarrow$, respectively; i.e., the representation in which H' is diagonal is simultaneously one where σ_Ω is diagonal. Hence, generally, near Γ, H' splits the Γ_6 states into pure spin-up and -down states referred to $\mathbf{\Omega}$. For $k \simeq 0$ the net polarization of the conduction band will be practically zero. However, for larger k a sizable polarization can occur as a result of the $\mathbf{k}$-dependent hybridization of valence states into the conduction bands which induces a mixing of different spins.[4] Electrons having the same energy E (see Fig. 1) will occupy states with $\mathbf{k}_\uparrow \neq \mathbf{k}_\downarrow$ and thus the degree of hybridization for each band is different. Therefore a net nonzero spin polarization results. In a photoemission experiment like the present, the *emitted* electrons do carry information about hybridization, but its observation will be strongly affected by transport and surface-transmission effects. In a crude model one may try to account for these by introducing weight factors proportional to the density of states $N(E,\mathbf{k})$. If we take $N(E,\mathbf{k}) \propto (dE/dk)^{-1}$, it follows from Fig. 1 that $N(E,\mathbf{k}_\uparrow) \neq N(E,\mathbf{k}_\downarrow)$. Hence, a nonzero polarization results, even in the absence of hybridization.

In the present paper we give evidence of this spontaneous spin polarization. The experiment consists of the measurement of the magnitude and orientation of the polarization $\mathbf{P}_\Omega$ of electrons photoemitted from negative-electron-affinity GaAs(110). Negative electron affinity established by the usual technique of Cs and O_2 surface coverage. The measurements were done with the experimental arrangement by Riechert *et al.*[6] Electrons are excited into the conduction bands by linearly polarized or "unpolarized" light (rotation of the linear polarizer in order to randomize the residual circular polarization at the view ports where the light enters the UHV apparatus). Therefore, the *optical* spin orientation of the photoelectrons can be neglected. The exciting radiation was $h\nu = 1.5$ eV ($E_g = 1.43$ eV at room temperture) so that only low-energy electrons are injected into the band-bending region. Here they are accelerated towards the vacuum and emitted with a mean $\mathbf{k}$ along [110]. For this momentum direction $\mathbf{P}_\Omega$ is along $[\bar{1}10]$; i.e., it is parallel to the crystal surface. The spin polarization is measured by Mott scattering. Originally, our setup was conceived to measure the component of the polarization *perpendicular* to the emitting surface. In order to measure this new effect we thus had to rotate the polarization of the emitted electrons by 90°, by means of a solenoid, before injection into the Mott detector. With rotation of the crystal about its normal by an angle ϕ the measured polarization thus follows a sinelike curve: $P_\Omega^y = P_\Omega \sin(\phi - \eta)$, where η is the angle between $\mathbf{P}_\Omega$ and the [110] crystal axis in the plane of the crystal. Figure 2 shows a typical P_Ω^y vs ϕ measurement.

Figure 3 shows the results of several measurements of P_Ω as a function of the electron energy E above the bottom of the Γ_6 conduction band. E is taken to be equal to the band bending δ at the surface of GaAs. This is the energy gained by electrons which ballistically cross the surface-depletion region towards the vacuum. δ is determined from a measurement of the precession angle θ_p[6,7] of the spin-polarization vector of conduction electrons optically oriented by circularly

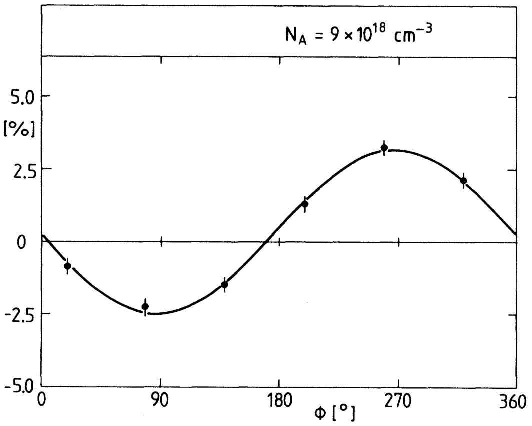

FIG. 2. Typical measurement of the spontaneous spin polarization as a function of the azimuthal angle between the $[1\bar{1}0]$ crystal direction and the momentum of the electrons entering the spin analyzer. The polarization component $P_\Omega^x(\phi)$ follows a sinelike curve whose phase allows us to determine the orientation of $\mathbf{P}_\Omega$ in the crystal. This result shows that $\mathbf{P}_\Omega$ is oriented parallel to the $[\bar{1}10]$ direction for $\mathbf{k} \parallel [110]$ as expected from the calculations shown in Fig. 3.

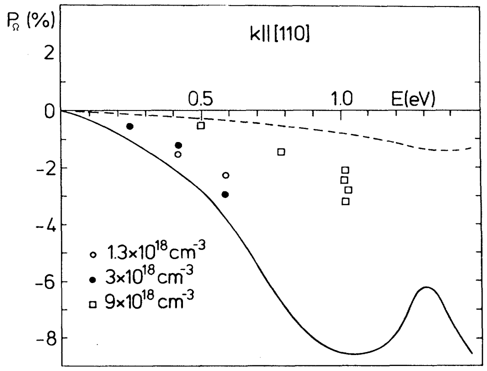

FIG. 3. P_Ω vs energy of the electrons with respect to the bottom of the conduction band. Dashed line: "band" polarization calculated by relativistic linear-muffin-tin-orbital method taking into account hybridization effects only. Solid line: polarization calculated by including also the density-of-states factors. The energy of the experimental points is taken as equal to the surfaces band bending measured *in situ* (see text).

polarized light. Those measurements serve at the same time to find the $[1\bar{1}0]$ axis of the crystal.[6] Different values of δ are obtained in samples with doping levels in the range $(1.3\text{–}9)\times10^{18}$ cm^{-3} and with different surface treatments (e.g., ion-bombardment, annealing, etc.; cf. Ref. 7). Figure 3 also shows the polarization calculated for $\mathbf{k} \parallel [110]$ from the energy bands of GaAs obtained from a linear-muffin-tin orbital calculation using the local-density approximation, but with the conduction states adjusted by means of delta-function-like external potentials.[8] The dashed line shows the band polarization (due to hybridization only); the solid line shows the polarization calculated by inclusion also of the density of states (DOS). Theory and experiment agree in the sign. The magnitude of $\mathbf{P}_\Omega$ is several times larger than that expected from hybridization showing that DOS factors also play an important role. Still the polarization is not as large as expected from the DOS. This is perhaps not surprising if we consider that a proper matching of the crystal wave functions does not fully restore[9] the DOS factors (which one might intuitively introduce in a simplified description). Additionally the experiment samples a finite distribution of wave vectors about the mean $\mathbf{k} \parallel [110]$.

These results show a new way of producing spin-polarized electrons from crystals without inversion symmetry by use of linearly polarized light. Higher polarization values are expected in other III-V compounds, where a larger spin splitting is expected for larger spin-orbit interactions and smaller band gaps than those of GaAs.[10,11] We finally note that for energies E_{crit} near the critical point in the Γ-*K* direction (see Fig. 1) electrons emitted from only one of the spin-split bands could be analyzed, implying that electrons of high transverse polarization can be emitted.

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