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Light-induced drift of electrons in metals

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Photoexcitation of electrons in solids is, in general, velocity selective. It is accompanied by a net current provided that the electron mobilities differ for excited and ground states. This effect considered earlier for intersubband transitions in semiconductors is extended to metals. Photoinduced currents in the range 0.1–100 μA are predicted for intensities of order 1 MW/cm^2. Experimental evidence for this effect is presented in the form of spatially asymmetric photoemission from rough silver films.

In the early 1970s Grinberg et al. [1] showed that intersubband transitions in semiconductors can produce a radiation-induced current sufficiently large that the change in electron momentum is of the order of the electron momentum itself rather than the photon momentum. They have dubbed this phenomenon the resonant photon drag effect (PDE). The PDE occurs when the charge mobilities differ for excited and ground bands so that, under resonant conditions [2], the balance between the four radiation-induced partial currents (two oppositely directed currents in each of the subbands) is destroyed and a large net current arises. A similar effect for transitions between parallel subbands in quantum wells, referred to as light-induced drift (LID), was suggested by Dykhne et al. [3], developed further by Luryi and Grinberg [4] and, later, by Stockman et al. [5]. The effect was observed experimentally by Wieck et al. in 1990 [6]. LID of electrons by transitions between Landau subbands in a semiconductor in a magnetic field was considered by Skok and Shalagin [7] and observed by Kravchenko et al. [7]. A related phenomenon, LID of gases, was first suggested by Gel'mukhanov and Shalagin [8] and observed by Antsygin et al. in 1979 [8]. More recent work on LID in gases has been reviewed by Werij and Woerdman [9]. In spite of the fact that these effects have peculiarities unique to each of the various systems, the underlying concept is similar in all cases. Accordingly the terms "resonant PDE" and "LID" are equally descriptive and we will arbitrarily use the latter.

In this Letter we extend previous treatments of LID to include electrons in metals and present experimental evidence for the effect.

In order to emphasize the essential physics we begin by considering a perfectly free electron metal with dispersion relations $E_1(\boldsymbol{k})=(\hbar^2/2m)\boldsymbol{k}^2$ and $E_2^{\pm}(\boldsymbol{k})=(\hbar^2/2m)(\boldsymbol{k}\pm\boldsymbol{G})^2$ corresponding to the initial and final bands, 1 and 2, respectively. $\pm\boldsymbol{G}$ are the reciprocal-lattice vectors which generate the 2d band. For metals, in general, the bands are not parallel even for equal masses in the two bands. Since $\boldsymbol{k}'=\boldsymbol{k}+\boldsymbol{q}$, $E_2(\boldsymbol{k}')=E_1(\boldsymbol{k})+\hbar\omega$, transitions between these two nonparallel bands at photon energy $\hbar\omega$ and momentum $\hbar\boldsymbol{q}$ are restricted to the surface of the constant interband energy (fig. 1; $\boldsymbol{q}\parallel\boldsymbol{G}$; k, $G\gg q$) given by

$$S(\boldsymbol{k})=E_2(\boldsymbol{k})-E_1(\boldsymbol{k})-\hbar\omega'$$
$$=(\hbar^2/2m)(\pm 2\boldsymbol{k}\cdot\boldsymbol{G}+G^2)-(\hbar\omega\pm qv_p)=0,$$

where ω' takes into account the Doppler shift of the frequency, $\omega'=\omega-\boldsymbol{q}\cdot\boldsymbol{v}_k=\omega\pm qv_p$, for an electron with velocity $\boldsymbol{v}_k=\hbar^{-1}\nabla_k E(\boldsymbol{k})$. The minus and plus signs in ω' correspond, respectively, to electrons moving with and against the light. For a free electron metal

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$\boldsymbol{v}_k=(\hbar/m)\boldsymbol{k}$ and $v_p=(\hbar/m)\ k_p$, where $v_p\equiv(\boldsymbol{v}_q)_p=\boldsymbol{v}_k\cdot\boldsymbol{q}/q$, $k_p\equiv\boldsymbol{k}\cdot\boldsymbol{q}/q$. The surfaces $E_1(\boldsymbol{k})$ and $E_2(\boldsymbol{k})$ are spheres indicated, respectively, by the circles i and f, f′ in fig. 1. The states between which interband transitions can occur are represented by disks in momentum space whose diameters are given by the lines AB and A′B′ in fig. 1. The crucial point for LID is that the distances of the centres of these disks are not equal with respect to the origin of momentum space owing to the Doppler effect (cf. fig. 1).

Illuminating the system results in transitions that populate the upper and deplete the lower bands slightly. However, this process will, in general, be unequal for electrons travelling along and against the surface projection of $\boldsymbol{k}$, since they involve electrons of somewhat different momenta. This leads to asymmetrical forms of the (nonequilibrium) momentum distribution of electron population $N_i(\boldsymbol{k})$ in the ith band ($i=1,2$). This implies that, upon illumination, nonzero currents of excited and unexcited electrons will be set up in the system:

$$\boldsymbol{j}_i=e\int \boldsymbol{v}_k N_i(\boldsymbol{k})\ \mathrm{d}\boldsymbol{k}\,, \tag{1}$$

where e is the electron charge.

The force, $\boldsymbol{F}$, that operates on the electrons immediately upon establishing the currents $\boldsymbol{j}_1$ and $\boldsymbol{j}_2$ will be of the form $\boldsymbol{F}=-\nu_1 m_1\boldsymbol{j}_1/e-\nu_2 m_2\boldsymbol{j}_2/e$; where ν_i and $\boldsymbol{m}_i$ are the collision frequencies, or more generally, scattering rates, and the effective masses of the electron in the ith band, respectively. When steady state is established, and assuming a closed circuit, $\boldsymbol{F}$ becomes zero and, since the total current $\boldsymbol{J}=\boldsymbol{j}_1+\boldsymbol{j}_2$, one obtains $\boldsymbol{J}=\boldsymbol{j}_2(\nu_1 m_1-\nu_2 m_2)/\nu_1 m_1$. Thus, a nonzero value of $\boldsymbol{J}$ is expected only when $\nu_1 m_1\neq\nu_2 m_2$.

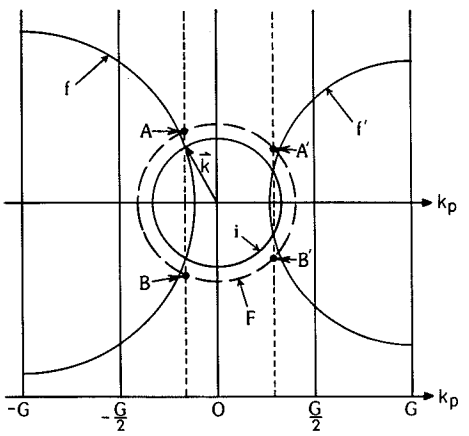

Fig. 1. Two-dimensional slice through momentum space taking the projection of the electron momentum upon the wavevector (directed along the reciprocal-lattice vector $\boldsymbol{G}$) of light as one of its coordinates. F indicates the Fermi sphere.

Note that in spite of the fact that the Doppler effect, critical for LID, depends on the photon momentum $\boldsymbol{q}$, neither the average magnitude of the acquired electron momentum nor its sign have anything in common with the momentum of the absorbed photons. The force $\boldsymbol{F}$ acting on the electrons by the lattice and the equal but opposite force operating on the lattice are due to the entropy transfer between photons and electrons.

The momentum distribution of excited electrons $N_2(\boldsymbol{k})$ is proportional to

$$N_2(\boldsymbol{k},\omega')\propto \mathrm{Im}\frac{|V_{12}(\boldsymbol{k})|^2}{E_2(\boldsymbol{k})-E_1(\boldsymbol{k})-\hbar\omega'+\mathrm{i}\Gamma}, \tag{2}$$

where $V_{12}(\boldsymbol{k})$ is the matrix element associated with the transition and Γ is its width. Referring to fig. 1, electron excitation takes place when the electron energy, E, lies within energy interval $E_{\min}<E<E_F$. The minimum initial energy, $E_{\min}$, corresponds to the situation where the spheres i and f (or f′) just touch. This energy is readily evaluated and, under the condition $\hbar q v_p\ll E_{\min}$, is given by $E_{\min}=(\hbar\omega-E_G)^2/4E_G$, where $E_G=G^2\hbar^2/2m$ ($m_1=m_2$). Taking into account the band gap at the zone boundaries, one obtains [10]: $E_{\min}=\frac{1}{2}mv_0^2=\frac{1}{4}[(\hbar\omega-E_G)^2-4|V_G|^2]/E_G$, where V_G is the Gth Fourier component of the pseudopotential. By considering the band gap we have, in principle, moved from a perfectly free electron to a nearly free electron model. As is apparent in fig. 1, the velocity v_0 is equal (to within accuracy $\hbar q v_0/E_{\min}$) to the velocity projection v_p of the excited electrons.

If homogeneous broadening dominates ($\Gamma\gg qv_0$), one can expand N_2 in a Taylor series and keep only first order terms in $\boldsymbol{q}\cdot\boldsymbol{v}_p$,

$$N_2(\boldsymbol{k},\omega')\approx N_2(\boldsymbol{k},\omega)-\nabla_\omega N_2(\boldsymbol{k},\omega)\boldsymbol{q}\cdot\boldsymbol{v}_k\,.$$

Only the second term in this expansion contributes nonvanishingly to the current

$$\boldsymbol{J}=-\frac{\boldsymbol{q}}{q}\frac{\nu_1 m_1-\nu_2 m_2}{\nu_1 m_1}\frac{qv_0}{\Gamma_2+\nu_2}ev_0\frac{I}{\hbar c}\frac{\partial\epsilon''}{\partial\omega}. \tag{3}$$

In obtaining (3) we assumed that the density of excited electrons $N_2\equiv\int N_2(\boldsymbol{k})\,\mathrm{d}\boldsymbol{k}$ under steady-state conditions is given by $N_2=\epsilon''I/\hbar c\Gamma_2$, where $\epsilon=\epsilon'+\mathrm{i}\epsilon''$ is the interband contribution to the dielectric function, Γ_2 is the relaxation rate from the second to the first band, c is the speed of light and I is the local light intensity. The factor $\Gamma_2+\nu_2$ in the denominator in (3) gives the momentum total relaxation of the excited electrons due to both the collisions keeping the electron in the second band and the inelastic relaxation to the first band.

For nearly free electrons the interband contribution to the imaginary part of the dielectric constant takes the form [10]

$$\epsilon''=\alpha\omega^{-4}(1-\gamma)^{-1}(E_\mathrm{F}-E_\mathrm{min}),$$

where $\alpha=\frac{2}{3}(e^2/\hbar^4)G|V_G|^2$, and $\gamma=1-[(\hbar\omega)^2-4|V_G|^2]^{1/2}/\hbar\omega$. Substituting this into (3) and using the expression for E_min, one obtains (when $m_1=m_2$)

$$\boldsymbol{J}=-\frac{\boldsymbol{q}}{q}\frac{\nu_2-\nu_1}{\nu_1}ev_0\frac{qv_0}{\Gamma_2+\nu_2}\frac{\epsilon''I/\hbar c}{E_\mathrm{F}/\hbar-E_\mathrm{min}/\hbar}\Phi_\mathrm{h}(\omega), \tag{4}$$

where the frequency dependent function $\Phi_\mathrm{h}(\omega)$ has the form

$$\Phi_\mathrm{h}(\omega)=4\frac{E_\mathrm{F}-E_\mathrm{min}}{\hbar\omega}\left(1+\frac{|V_G|^2}{(\hbar\omega)^2-4|V_G|^2}\right)+\hbar\omega/2E_G-\tfrac{1}{2}. \tag{5}$$

The singularity in $\Phi_\mathrm{h}(\omega)$ at $\hbar\omega\to 2V_G$ can be eliminated by including a damping constant Γ in the denominator. The maximum current value

$$\boldsymbol{J}_\mathrm{max}\sim\frac{\nu_1-\nu_2}{\nu_1}\frac{\Gamma_2}{\Gamma_2+\nu_2}\frac{qv_0}{\Gamma}N_2ev_0$$

is reached at a photon energy close to the energy gap $\hbar\omega-2|V_G|\approx\hbar\Gamma$. With a further increase of the laser frequency, the LID current decreases and then changes its sign. Under optimum conditions the difference between partial currents corresponds to an acquired macroscopic momentum per electron of the order of k rather than q.

The photon momentum affects electron excitations in two ways. First, momentum and energy conservation lead to the Doppler relation and determine the group velocity of electrons excited by photons with frequency ω. As a result of the velocity selective excitation the acquired macroscopic velocity, corresponding to the directed motion of the electrons as a whole, is determined by the factor $v_0[\partial_\omega\epsilon''(qv_0)]$ (see (3)), which, under the optimum condition $\hbar\omega-2|V_G|\approx\hbar\Gamma$ and $\partial_\omega\epsilon''\sim\epsilon''/\Gamma$, gives $v_0(qv_0/\Gamma)$ or v_0, if $\Gamma\sim qv_0$. Second, the nonverticality of the transitions leads to an additional contribution to the macroscopic velocity determined by the factor $\epsilon''(\hbar q/m)$ which is usually associated with the radiation pressure. It follows from the exact solution that, unless the conditions are very far from optimum when $\partial_\omega\epsilon''\sim\epsilon''/\omega$, this term can be neglected.

When the transition is inhomogeneously broadened ($\Gamma\ll kv_0$), substituting (2) into (1), one finds

$$\boldsymbol{j}_2\propto\int\boldsymbol{v}_k|V_{12}(\boldsymbol{k})|^2\times|\nabla_k[E_2(\boldsymbol{k})-E_1(\boldsymbol{k})-\hbar\omega']|^{-1}f^{(0)}(\boldsymbol{k})\,\mathrm{d}S,$$

where $f^{(0)}$ is the equilibrium distribution function. After integrating over the surfaces of the constant interband energy, one obtains

$$\boldsymbol{J}=-\frac{\boldsymbol{q}}{q}\frac{\nu_1 m_1-\nu_2 m_2}{\nu_1 m_1}ev_0\frac{I/\hbar c}{\Gamma_2+\nu_2}q\frac{\partial}{\partial\omega}(\epsilon''v_0). \tag{6}$$

Substituting the expression for ϵ'' used above into (6) a formula for the total electron current is obtained similar to eq. (4) in form but with $\Phi_\mathrm{inh}(\omega)$ replacing $\Phi_\mathrm{h}(\omega)$, where

$$\Phi_\mathrm{inh}(\omega)=\Phi_\mathrm{h}(\omega)+\frac{1}{4}\frac{E_\mathrm{F}-E_\mathrm{min}}{E_\mathrm{min}}\left(1-\frac{\hbar\omega}{E_G}\right). \tag{7}$$

Using (4) one estimates a current density $\boldsymbol{J}\sim(10^{-7}\text{–}10^{-4})I$ A/W. This would result in an open circuit potential difference $\Delta U=L|\boldsymbol{J}|/Ne\mu$ across the illuminated sample of length L (where N is the electron density and μ is the effective electron mobility) corresponding to $\Delta U\sim 0.1$–100 μV for $I=1$ MW/cm^2, $L=1$ cm and typical values of N and electron mobility $\mu\sim e/mv^{-1}\approx 10^4$ (CGSE). The optical skin depth of metals is usually of the order of 10^2 Å. Accordingly, the current is expected to be in the range 0.1–100 μA. The predicted magnitude of the effect

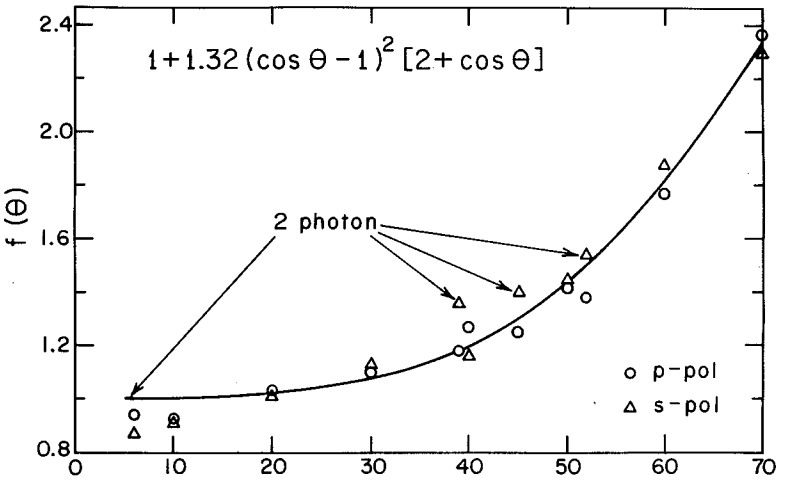

Fig. 2. Measured (points) and calculated angle of incidence function of the LID contribution to the total photoemission from a rough silver surface. Points not marked two-photon were obtained with one-photon excitation.

is large enough that measurable currents are possible even with cw radiation fluxes in the 1 W/cm^2 range.

It is perhaps worth emphasizing that the validity of expressions (3) and (6) is not restricted by the assumption of the nearly free electron model [#1] although, of course, eqs. (4) and (7), and the order of magnitude estimates, are. LID is, therefore, expected to be a rather universal phenomenon observable in a broad range of materials.

We have two observations that provide strong, indirect evidence for LID.

Stuckless and Moskovits [11] reported that the total laser-induced photoemission yield as a function of angle of incidence, for a cold deposited, and therefore rough, silver film was well represented by the expressions $Y_{s,p} \propto (1-R_{s,p}) f(\theta)$, for one-photon and $Y_{s,p} \propto (1-R_{s,p})^2 f(\theta)$, for two-photon excitation (s and p imply TE and TM polarizations, respectively, and R is the total reflectance calculated from known optical constants of silver). $f(\theta)$ calculated from all of these results was independent of the state of polarization of the incident light, within experimental error. This, plus the fact that on going from a one-photon to a two-photon experiment the angle of incidence dependence of the yield did not change from $f(\theta)$ to $f^2(\theta)$, while the absorptive part of the expression for the yield did change from $1-R$ to $(1-R)^2$, leads us to conclude that the $f(\theta)$ is a consequence of the LID.

For small metal surface features the LID current would result in an additional electron current crossing lateral metal–vacuum interfaces provided that the size of the feature is not much larger than the bulk electronic mean free path. Only microscopically rough surfaces are, therefore, expected to manifest this effect noticeably (for smooth films $f(\theta)$ was unity for all values of θ). One can show, in fact, that, for a hemispherical surface boss, only a fraction $p(\theta) = \frac{1}{2}(1-\cos\theta)^2(2+\cos\theta)$ of the electrons moving along $\boldsymbol{q}$, due to LID, can reach the metal–vacuum interface. One, therefore, expects that $f(\theta) = 1+\kappa p(\theta)$, where κ is of the order of the ratio of the probabilities that a hot electron reaches the surface by LID as opposed to the normal conduction processes. Figure 2 shows that a function, $f(\theta)$, of this form (solid line) fits the experimental data very well.

A series of experiments were performed in order to look for asymmetric photoemission directly. Briefly, unpolarized light from an XeCl excimer laser (4.0 eV) was divided into two beams arranged so that each passes through an appropriate window of a UHV chamber and impinges, with equal angles of

[#1] Formulas (3) and (6) can be obtained directly from the kinetic equation for the one-particle density matrix with collision integral of the Boltzmann type. Indeed, the effective velocity v_0, in general, is determined via the corresponding moment of a velocity distribution of the excitation cross section and may be specified in all particular cases.

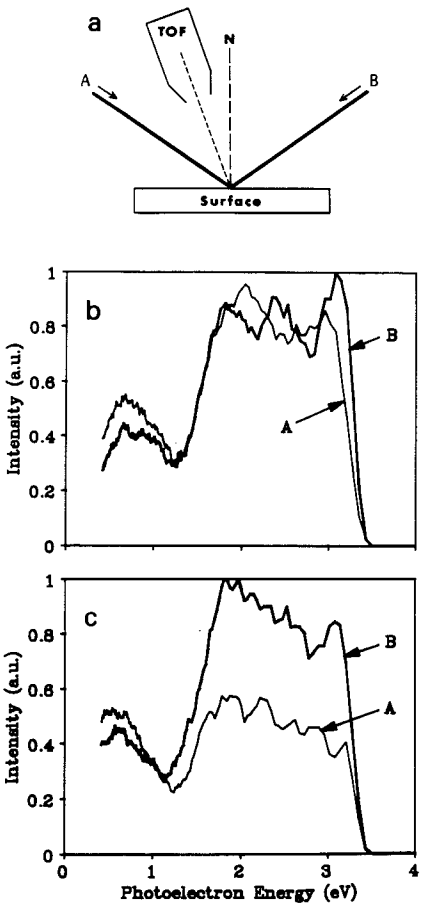

Fig. 3. (a) Schematic of the experimental geometry. A and B indicate the two approach directions of the laser. N and TOF denote the surface normal and the time-of-flight analyzer respectively. (b) Two-photon electron spectrum of a smooth silver film. (c) As in (b) but for a rough silver film. Although plotted in arbitrary units, the vertical scales for the two sets of spectra are approximately equal.

incidence (55°) but from opposite directions, on a silver surface deposited under UHV conditions. A time-of-flight electron energy analyzer was situated asymmetrically, at an angle of 20° with respect to the surface normal as in fig. 3a. The experiment was carried out both for smooth and rough films. The intensities of the two rays were made equal within the experimental error and checked by measuring the total photoemission yields using the sample recharge current.

The spectra in figs. 3b and 3c show the two-photon photoemission spectra obtained, respectively, from a smooth silver film deposited at room temperature, and from a rough film deposited at 30 K. The films were examined at their respective deposition temperatures. Our results imply a greater propensity for photoemission in the direction of the wavevector of the light only for the rough films, in agreement with the theory when $\nu_2 > \nu_1$ and on recognizing that photoemission probes the current of excited electrons only.

The possibility that the effect was a result of an asymmetry of the roughness features themselves with respect to the metal surface normal was investigated and eliminated by purposely growing rough surfaces with oblique surface features by depositing silver obliquely ($\pm 45°$) on the cooled substrate. The photoemission asymmetry did not depend significantly on the mode of preparation of the surface. (The deposition direction in the experiments reported in fig. 3 was normal to the surface.)

One should note that the experimental setup, intrinsically, breaks symmetry with respect to the surface normal so that even in the absence of LID asymmetric photoemission might be possible. The failure to observe asymmetric photoemission for smooth films, the insensitivity of the effect to the direction of the orientation of the roughness features, and the unusual angle of incidence effect on the *total* photoyield reported in ref. [11] all imply that this intrinsic symmetry breaking is not responsible for our observations.

We conclude, therefore, that the asymmetry in the angular distribution of photoelectrons with respect to the surface normal observed for rough silver films is a manifestation of light-induced drift.

The possible device and switching applications of LID have not escaped our notice.

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