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MULTIPHOTON PHOTOEMISSION FROM SOLID STATE,
WITH SPECIAL REGARDS TO METALS

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MULTIPHOTON PHOTOEMISSION FROM SOLID STATE, WITH SPECIAL REGARDS TO METALS

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ABSTRACT

The review of the theories and the experiments carried out in the field of the laser induced multiphoton photoelectron emission of solids is given. The article is dealing with the possibilities and realisations of observation of the multiphoton photoeffect and the optical tunnel emission in a Richardson-emission background using giant pulses and ultrashort laser pulses of extreme high intensities and extreme short durations. The literature of the related latest theoretical and the experimental results are summarized.

PESIONE

Дается обзор теоретических и экспериментальных работ, выполненных в области многофотонной фотоэлектрической эмиссии в твердых телах под действием лазерного излучения. В статье рассматриваются возможности наблюдения и методы исследования многофотонного фотоэффекта и оптической туннельной эмиссии на фоне эмиссии Ричардсона, используя гигантские импульсы и ультракороткие лазерные импульсы экстремальной высокой интенсивности и экстремально короткой длительности. Собрана литература по последним теоретическим и экспериментальным исследованиям, связанным с этими вопросами.

KIVONAT

A szilárd testekben lézersugárzás hatására fellépő többfotonos fotoelektron-emisszió problémakörének elméleti és kisérleti munkáira vonat-kozóan áttekintő összefoglalást adunk. A cikk az óriás lézerimpulzusok, illetve az extrém nagy intenzitásu és extrém rövid időtartamu ultrarövid /mode-locking/ lézerimpulzusok által kiváltott többfotonos fotoeffektus és optikai tunnelemisszió Richardson-háttér mellett történő megfigyelésének lehetőségeit és a kisérletek megvalósitását tárgyalja. Összefoglaljuk a legujabb elméletek és kisérleti eredmények irodalmát.

I. Introduction

II. Review of theoretical work

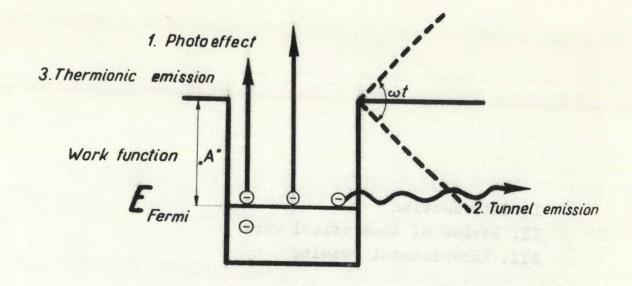
III. Experimental results

I. Introduction

When the high-intensity oscillating electromagnetic field of a laser beam interacts with the surface of a metal or some other solid, electron emission can be elicited. This process can take place in three ways:

- 1. by photoelectric effect;
- 2. by tunnel-emission due to the oscillating electric field;
- 3. by thermionic emission.

The three processes can be illustrated on the simplified rectangular potential valley of metals. (See Fig. 1.)



Electron emission from metals

F I G. 1.

In this lecture we shall be concerned primarily with the direct photon-electron interactions involved in the 1. and 2. processes. However we cannot ignore althogether thermionic emission resulting from indirect photon-electron interactions, because this generally occurs along with the first two phenomena and from that point of view represents a background to be eliminated.

Before the advent of lasers photoemission was known only in the form of the linear photoelectric effect characterised by Einstein's equation $h\nu=\,A\,+\,\frac{1}{2}\,\,\mathrm{m}\,\,v^2\,\,, \ \, \text{and by the linear relation} \,\,\,j\sim I$ between photoelectric current $\,\,j\,$ and the light intensity $\,\,I_{\bullet}$

Tunnel emission had been observed only in a static field or in low radiofrequency oscillating fields at high field strengths of about 10⁶ V/cm.

The discovery of the laser, however, brought the opportunity to realise these processes in new forms. The perturbations which can be achieved with the very high field strength produced by a laser beam may reach the binding strength of the electrons in metals. In these circumstances the photoelectric effect is produced by simultaneous absorption of n quanta. Einstein's equation and the linearity relation obtained from simple perturbation theory are no longer valid and in their place we have to use equation $nhv = A + \frac{1}{2}m v^2$ and the nonlinearity relation $j \sim I^n$ obtained from higher order approximations of perturbation theory, where $n = \left[\frac{A}{hv} + 1\right]_{ent}$ is the order of nonlinearity.

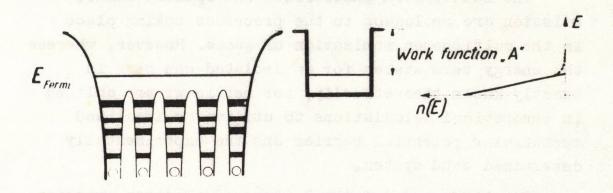
With extremely large perturbations there is strong deformation of the potential barrier and optical tunnel emission can be expected to occur, at sufficiently low frequencies, if electrons can pass the narrowed potential barrier during a half period of the laser light.

The multiphoton photoeffect and optical tunnel emission are analogues to the processes taking place in the multiphoton ionisation of gases. However, whereas the energy term system for an isolated gas atom is exactly known theoretically, for metals we are obliged in theoretical calculations to utilise an idealised rectangular potential barrier and the experimentally determined band system,

Detailed study of the 3. type of electron emission, Richardson emission, due to the heating of the metal by the absorbed light, enables us to take into account or to eliminate it in experiments on the pure photon-electron interaction, especially in picosecond time durations.

II. Review of theoretical background

For the formulation of the theoretical calculations we start from a simplified model of the electronic structure of metals. In the periodic field of a crystal lattice there exists a potential distribution with allowed and forbidden energy bands.



At the boundary surface of the metal the potential energy of an electron asymptotically approaches zero value and therefore the surface represents a potential barrier for electrons freely moving in the upper, only partially filled conduction band. We consider the conduction band as being symply a rectangular potential valley of Fermi energy E_F having a work function of value "A" and with the well known energy distribution function.

According to Sommerfeld [1], we may regard the conduction electrons as comprising a free electron gas system. How then can photoelectric effect arise in this model? Tamm and Schubin [2] have shown that Sommerfeld type free electrons cannot absorb a photon because energy and momentum cannot both be conserved in such a process. For this the presence of a "third body" is necessary. There are two possibilities for photon absortion satisfying this condition. First an electron may absorb photons in the field of an abrupt potential jump /10 cm/ on the surface /the third body/: a process which depends only on the opticalfield strength component perpendicular to the surface and which for metals has a threshold energy of about 2 - 5 eV. This is the surface photoeffect. Alternatively, an electron bound to the periodic field of the lattice absorbs photons, in which case the lattice represents the "third body". This is the volume photoelectric effect; it does not depend on the polarisation of the optical field strength and it has a high threshold energy of 8-10 eV.

Theoretical and experimental work shows in fact that apart from certain special cases the photoelectric effect of metals investigated at laser frequencies is purely a surface photoelectric effect. Let us, then, briefly review the theoretical work relating to the surface multiphoton photoeffect. (An excellent review of these theories is given by Barashev [3]).

Consider a plane monochromatic light wave of frequency \vee impinging on the plane boundary of a metal of work function x, for which $n-1<\frac{A}{h}< n$, where n is an integer. The metal-vacuum boundary plane coincides with the xy coordinate plane, and the metal occupies the half space x<0. Assuming a definit potential model for the conduction electrons of the metal, we want to find the photoemission current from the metal into the vacuum.

This problem can be broken down into two parts:

a/ First the x component of the partial electric current density must be determined for x+∞:

$$j_x = \frac{ieh}{2m} (\Psi \nabla \Psi^* - \Psi^* \nabla \Psi) - \frac{e^2}{mc} \vec{A}$$

b/ In the second step the total photocurrent j is calculated by integrating j_x over the momenta of the emitted electrons p and over the electron states in the metal: $j = \int j_x (p, v) W_F(p) dp$, where $W_F(p)$ is the Fermi - Dirac distribution. In the expression j_x , A is the vector potential of the wave and Ψ satisfies the Schrödinger's equation /with Coulomb - gauge/.

$$i\hbar \frac{\partial \Psi}{\partial t} = \left(-\frac{\hbar}{2m} \nabla^2 + V(x) + \frac{i\hbar e}{mc} \vec{A} \nabla + \frac{e^2}{2mc^2} \vec{A}^2\right) \Psi$$

in which V(x) represents the potential barrier of the metal surface. The various theoretical treatments for calculating the partial current density $j_{\mathbf{x}}$ differ fundamentally in their approaches, i.e. in their

solution of the first part of the problem. We can broadly distinguish between two groups:

- 1/ Treatments based on higher order approximations of the perturbation theory; and
- 2/ Methods of calculation which dispense with perturbation, theory.

1/ The methods of the first group are essentially generalisations for higher order processes of the first-order perturbation calculation elaborated by Mitchell [4] for the linear photoeffect.

The feasibility of observing the two quantum surface photoeffects was first discussed in a paper by Makinson and Buckingham [5], and later by Smith [6] and Adawi [7] on the basis of the second order perturbation. The errors in Smith's calculations were corrected by Marinchuk [8], who has given the correct result. Starting from these precedents, Barashev [9] was able to find explicit formulas for the polarisation dependence of the two quantum surface photoeffects. Of methodological interest, we may mention here the sole work relating to the volume multiphoton photoeffect, the second-order theory of Bloch and its modification by Teich and Wolga [10] for Na metal.

In a detailed study by Brodsky and Gurevich [11] the generalisation of the so-called threshold production phenomena of quantum mechanics was applied to determine j_x . With this more general perturbation method they were able to take into account the final state interactions, a feature missing from former treatments both in the case where the field of the laser perturbation radiation is time dependent, and long-range time-independent Coulomb field of the cathode due to the emitted charges.

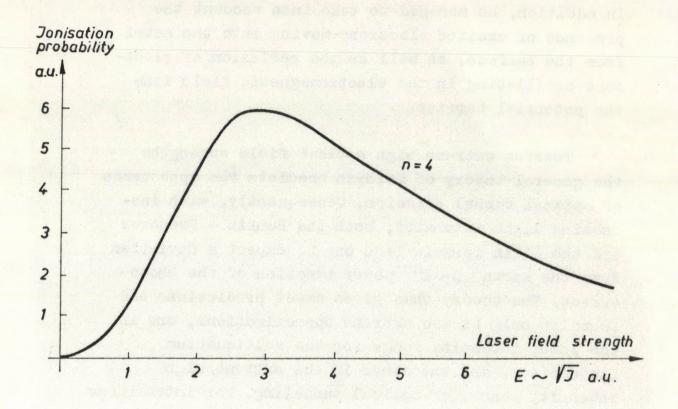
The results indicated that the dependence of the photocurrent on the light intensity is stronger than the usual nth order power dependence; the explicit formula, however, was not given.

Though application of perturbation theory can be relied upon to be successful in the case of second - and /to some extent/ third - order photoeffects for larger values of $n = \frac{A}{h\nu}$ the calculation difficulties increase to such an extent that this approach becomes impracticable.

2/ The second group of treatments tackles the problem of determining the current denisity without the use of the simple perturbation theory. The semiclassical theory of Keldysh [12] is valid for both gases and solids, and avoids the difficulties by dispensing with perturbation calculation but without making any foreign phenomenological preconditions, except for the assumption of a step-barrier for the potential form. This general theory has been applied to the photoeffect of metals by Bunkin and Fyodorov [13] . who calculated the probability of electron transition from a definite initial state in the metal, not to a stationary final state corresponding to the free motion of the electron, but to the "Keldysh final state". At relatively low laser intensities the theory describes the process of the nth order nonlinear photoeffect and was originally thought to be valid mainly for large n. Marinchuk [8] demonstrated, however, that this approach gives better results than the usual perturbation theory even for n = 2and n = 3. In a later re-elaboration of the Keldysh-Bunkin-Fyodorov method Silin [14] obtained a formula which is valid both of high and low values of n.

In addition, he managed to take into account the presence of excited electrons moving into the metal from the surface, as well as the reflexion of electrons oscillating in the electromagnetic field from the potential barrier.

Towards extreme high optical field strengths the general theory of Keldysh predicts the appearence of optical tunnel emission. Consequently, with increasing light intensity, both the Bunkin - Fyodorov and the Silin formula lead one to expect a deviation from the known j~In power function of the photoeffect. The theory thus gives exact predictions and formulas only at two extreme approximations, one in the lower intensity range for the multiquantum photoeffect, and the other in the extreme high intensity range for optical tunneling. For intensities corresponding to the transition between the two extremes there is no exact theoretical model except Silin's, which suggests a slower variation of the photocurrent j than power function j~In, though without giving an explicit formula .Similar results have been recently obtained for the decrease in the order of nonlinearity in the high intensity range by Reiss [15] using the momentum translational methode; who gave analitical formulae, too, for hydrogen atom, and not for metals. In Fig. 3. the dependence of the ionisation probability on the laser field strength E is plotted, following from his theory.



F I G. 3.

It should be evident that so far as theoretical investigation of multiphoton photoemission is concerned two essential problems are outstanding:

- 1/ The role of the coherence properties, and
- 2/ The role of the thermionic background.

As for the coherence, an amount of theoretical investigations describing the connection between multiphoton processes and the higher-order coherence properies of light have shown (see in the review paper [16] of Barashev) that the probability of the processes depends on a quantity $\langle E^{2n} \rangle$, whose link with the intensity $I \equiv \langle E^2 \rangle$ is determined by the relation $\langle E^{2n} \rangle = f_n \langle E^2 \rangle^n$, where f_n is the correlation function. The process can thus be described exactly with a knowledge of f_n , i.e. of the coherence properties. The general theory has been applied to the

nonlinear photoeffect by Teich and Wolga [17] and in a more detailed form, by Barashev [18] . These investigations are interesting in two respects. Firstly in the case of light with known statistics it is found possible to investigate the elementary nonlinear photoeffect. Conversely, with a multiphoton photodetector of known characteristics, the higher-order coherence properties of light should be determinable. Similarly to the case of linear photon statistics the photoeleectron distribution p/m/ and the light intensity distribution p/I/ can be determined from each other. The photoelectron distribution and the higher momentumfunctions were worked out by Barashev for the multiphoton photoeffect. Barashev also established that the yield can be varied by the well known factor n!, and that the form of the function p/m/ may be strongly distorted for a small variation of the cohorence properties of p/I/.

The second important problem I mentioned is the presence of thermionic emission, which may contribute a strong background at high intemsities. Detailed studies of this problem can be found in the books by Ready [19] and Anisimov [20]. Anisimov demonstrated that two kinds of emission can be expected: one due to heating of the whole crystal, and a second due to heating of the conduction electrons alone, which have only a very small a specific heat and hence have an important role in the case of ultrashort pulses.

There are two possibilites of eliminating, or at least reducing the thermal background. Bunkin and Prohorov [21] suggested that by reducing the duration of the light pulse and at the same time increasing of its intensity, the higher order multiphoton interaction can be made be predominant at the expense of the single-photon heating process. The second

possibility is offered when the light beam strikes
the cathode surface in grazing incidence. In this
case the emitting surface is automatically larger
and by using metal cathodes of high reflectance the
absorbed energy can be reduced considerably. These
two techniques are combined optimally in the experiments that have been carried out at the Central
Research Institute for Physics in Budapest.

III. Experimental results

The experimental work has succeeded in establishing the following evidence to prove the existence of the multiquantum photoeffect:

1/ The photcurrent depends only on the component of the optical field strength which is perpendicular to the cathode surface /Polarisation dependence/.

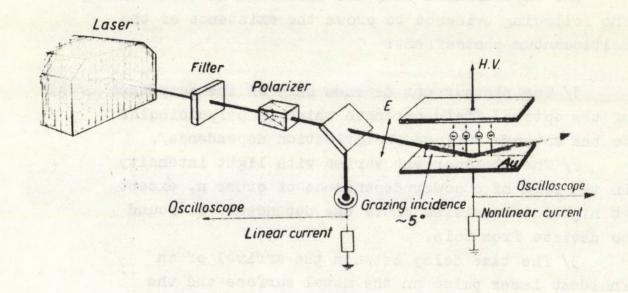
2/ The photocurrent varies with light intensity in the form of a power dependence of order n, except at higher intensities where the dependence is found to deviate from this.

3/ The time delay between the arrival of an incident laser pulse on the metal surface and the emission of the photoelectron pulse is zero. Only thermal electrons are delayed.

4/ Owing to the nonlinear response of the cathode, the duration of the photoemission pulse T is shorter than that of the exciting laser pulse T. With Gaussian pulse shapes $T = \frac{T}{n}$.

5/ The energy distribution of photoelectrons exhibits a maximum, while that of thermal electrons has an expenential form.

The various experimental set-ups used to study the multiquantum photoeffect are on the whole similar to one other and differ only slightly in details. (See Fig. 4)



F I G. 4.

The laser beam is passed through a variable attenuation and split into two parts, one part is directed to a linear photocell which is coupled to an oscilloscope for the linear detection /Determination of the duration of ultrashort pulses can be performed with a similar deviated beam in a TFF. system/. The other part of the beam is directed to the nonlinear metal cathode through a Glan-Thompson polariser and a lens. The multiquantum photocurrent pulse of the cathode is observed on a second oscilloscope. The cathode and the collecting electrode /or electron multiplier/ are situated in a closed vacuum system at a low pressure of 10^{-6} - 10^{-8} mmHg.

The measurement itself consists of simultaneously observing the signals of the linear and nonlinear detectors as a function of the different parameters: intensity, polarisation, pulse duration, etc.

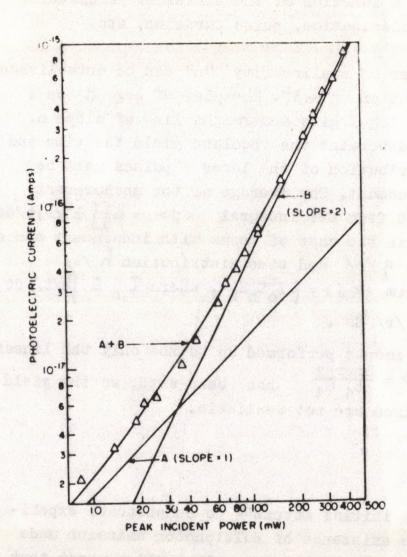
The order of nonlinearity "n" can be established from the relation $j \sim I^n$. The plot of log j as a function of log I give a straight line of slope n. In order to determine the absolute yield the time and surface distribution of the laser pulses must be taken into account. The average of the measurement is calculated from the integral $\langle j \rangle = \beta \int \int I^n/r, t/ds \ dt$,

which in the case of beams with indpendent space distribution g/r/ and time distribution h/t/ takes the form $\langle j \rangle = \beta I_o^n t_n S_n$, where $T_n \equiv \int h/t/ dt$ and $S_n \equiv \int g/r/ ds$.

In the experiments performed up to now only the linear average $\langle I \rangle = \frac{\text{Energy}}{t_1} S_1$ has been used, so the yield values obtained are not realistic.

Results

All the initial attempts to demonstrate experimentally the existence of multiphoton emission made use of relatively long laser pulses which meant that the process being sought was masked by strong thermionic emission Teich, Schroer, and Wolga [22] for instance examined the "j-I" intensity dependence of the emission of a Na cathode. (See Fig. 5.) At lower intensities the photoemission was linear due to the electrons of the Fermi tail, while towards higher intensities Richardson emission dominated, and only asymptotic statements could be given for the j~Iⁿ relation.

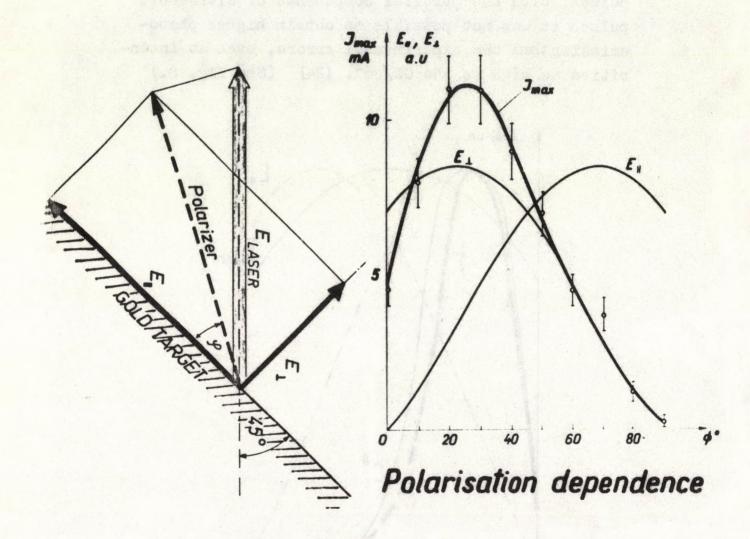


F I G. 5.

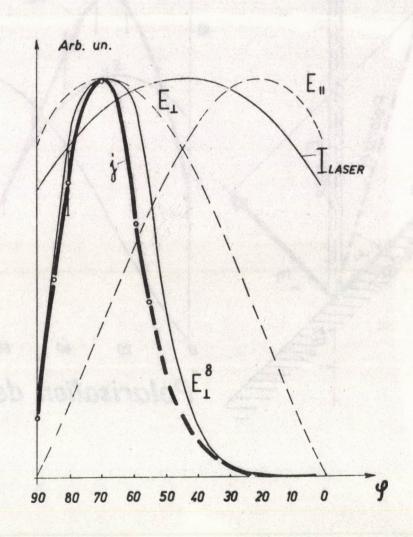
The first correct results were obtained in 1965 at Budapest and Shortly afterwards at Cornell University, in investigations of the third-order photoeffect of a gold cathode, It is these and the subsequent results that I want to deal with in the remaining part of my talk.

1/ Polarisation dependence

By varying the angle of polarisation of light with respect to the plane normal to the cathode surface, it was shown in Budapest (See Fig. 6. and Fig. 7.) that the photoemission depends only on the field component perpendicular to the cathode surface [23].

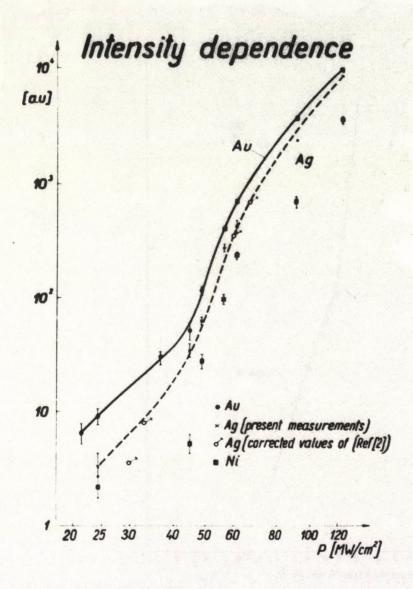


This is proof that the effect is a surface effect, in accordance with the calculation of Barashev [9]. The experiment was carried out with giant pulses of a ruby laser, of $T_c = 25$ nsec duration, k v = 1.8 eV and ~ 50 MW/cm² power trained on a gold cathode /A = 4.7 eV/ at grazing incidence. The polarisation dependence gives information on the role of the volume photoeffect and thermionic effect: when the field strength component was parallel to the surface, photoemission disappeared. Shortening the duration of the pulses, with the parallel components of ultrashort pulses it was not possible to obtain higher photoemission then the experimental errors, even at intensities as high as 10 GW/cm². [24] (See Fig. 8.)



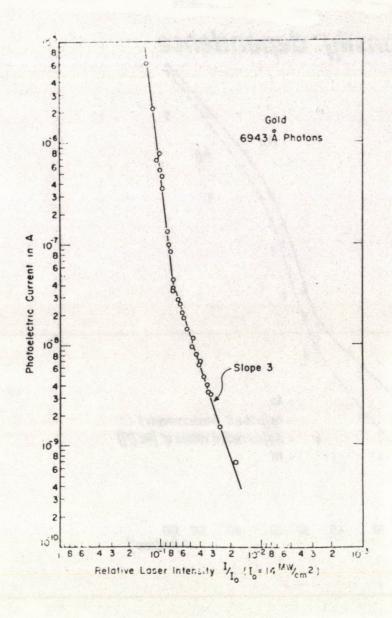
2/ Intensity dependence

Further experiments with giant pulse lasers showed [25] that the $j \sim I^n$ relation is valid only in the low intensity range i.e. at intensities 50 MW/cm² Towards higher intensities the slope of the curve was found to be greater than "n". This phenomenon is thought to be due mainly - if not entirely - to the occurence of Richardson-emission. (See Fig. 9.)



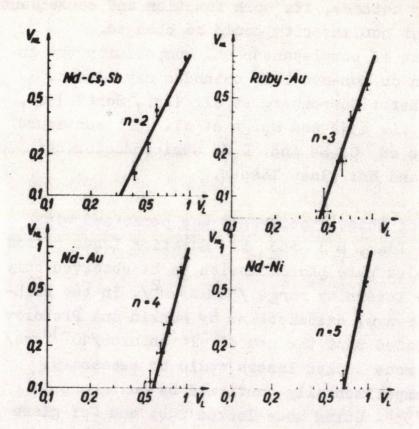
The experimental conditions and the errors, however do not exclude, a certain contribution from the final state interactions calculated by Brodsky and Gurevich [11], in this intensity range where $j = \alpha/I/I^n$.

These measurements were performed mostly in Budapest and by the Cornell University team of Logothetis, Hartman, [26] [27] (See Fig. 10.) Teich and Wolga [10] using Au, Ag, Ni, stainless steel and Na metal cathodes, with ruby and Nd: glass giant pulse lasers and gallium arsenide semiconductor lasers.



But similar results have been reported Korshunov at al. [28] for the multiquantum photoemission from a mercury cathode placed in solution. This latter group found that by varying the outer potential of the mercury cathode, its work function and consequently the order of nonlinearity could be changed. For the sake of completeness and comparision the investigation on non-metallic cathodes may also be mentioned here: Sonnenberg et al. [29], Soref [30], Görlich et al. [31] and Shiga et al. [32] continued experiments on C_{s3} Sb and K_3 Sb semiconductor cathodes with ruby and Nd: glass lasers.

Each of these experiments was performed with relatively long, μ s and ns duration laser pulses which enabled pure photoemission to be observed only in the low intensity range /50 MW/cm²/. In the high-intensity range, calculations by Bunkin and Prohorov [21] indicated that the use of ultrashort /10 $^{-12}$ sec/pulses of mode locked lasers would be necessary. This was experimentally confirmed by our group in Budapest [33]. Using mode-locked Ruby and Nd: glass lasers and Au, Ni and $C_{\rm S}$ Sb cathodes we found that the pure nonlinear photoeffect extended to the gigawatt intensity range. (See Fig. 11., where $\rm V_L$ is proportional to I and $\rm V_{NL}$ to j.)



Intensity dependence (Ultrashort pulses)

F I G. 11.

The results are summarised in the Table where the material of the cathode, the work function, the wavelength, the order of nonlinearity and the cathode efficiency j/I are given.

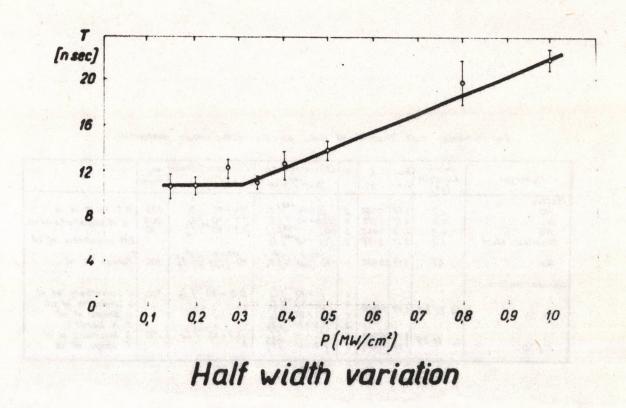
Experimental and theoretical data on the multiquantum photoeffect

Malerial	function (eV)	hw (eV)	λ (Å)	n	[Acm 3/Wcm 3		exp theor	Ref
Metals Na Au Au Stainless steel	5,0	3,57	3472 6943 3472	2 2 3 2	(8 * 10 -18) I. (23,5 * 10 -10) I. (10,2 * 10 -10) I. (4,2 * 10 -10) I. -10 (m) v) I.	(24 · 10 · 1) [. (12 · 10 · 1) [. (12 · 10 · 1) [. (12 · 10 · 1] [.	330 200 10	M.C. Teich et al E.M. Logothelis et al EM Logothelis et al.
Au Semiconductors	47	1,17	10600	4	(2 = 10 ⁻¹⁶) I. (6 = 10 ⁻¹⁷) I.	10 (cm v) 1.	100	Farkas et al H. Sonnenberg et al.
Cs, Sb	1,8 to 2.0			2 2 2 2	(2.1 = 10 -1) I. (4.2 = 10 -1) I.	(5.7 × 10 "5) I.	70	S. Imamura et al. F. Shiga et al. R. A. Seref S. Imamura et al.
K, Sb	2,2 to 2,9	1,17	10600	3	(1.6 - 10 - 1]	(5,7 = 10) 10	,0	F. Shiga et al.

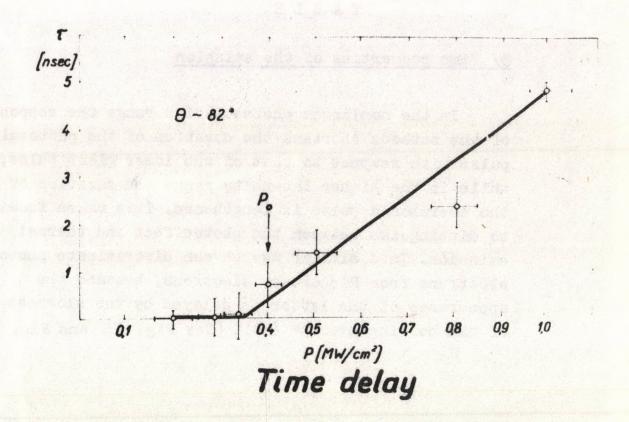
TABLE

3/ Time properties of the emission

In the nonlinear photoemission range the response of the cathode shortens the duration of the photoemissive pulse with respect to that of the laser giant pulse, while in the higher intensity range the duration of the thermionic pulse is lengthened. This makes feasible to distinguish between the photoeffect and thermal emission. In a similar way we can discriminate photoelectrons from Richardson electrons, because the appearance of the latter is delayed by the slowness of the heating process [34]. (See Fig. 12. and Fig. 13.)



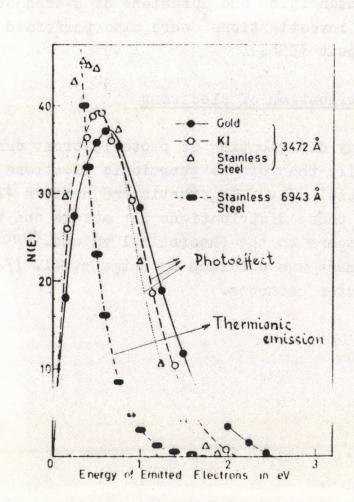
F I G. 12.



Both phenomena have lead to the 50 MW intensity value for the upper limit of the observations of photoeffect in accordance with the intensity dependence measurements, using nsec giant pluses. Because nonlinear detectation is sensitive to the density of photons and not the integral number of photons, nonlinear detectors can be used for the determination of the real intensities and durations of ultrashort pulses. These investigations were also performed by us in Budapest [35].

4/ Energy distribution of electrons

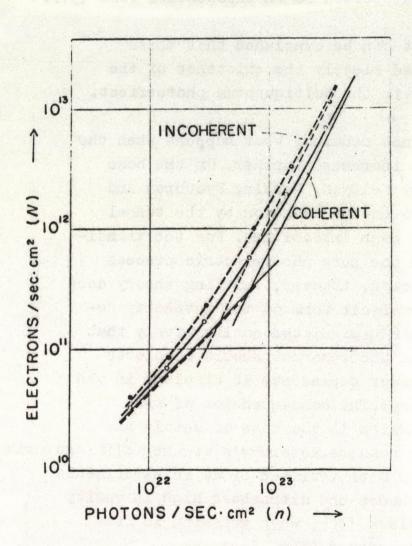
The energy distribution of photoelectrons exhibits a maximum, while that of the thermionic electrons is an exponential curve. Logothetis and Hartman [27] have measured these distributions for metals and their results correspond to the theoretical values. (See Fig. 14.) Similar agreement was obtained by Shiga et al. [32] for semiconductor cathodes.



F I G. 14.

5/ Statistical properties

Experimental verification of the prediction that the yield of multiquantum photoemission is a factor n! higher in the incohorent case than in the cohorent case has been provided by Shiga and Imamura for semiconductor cathode. [36] (See Fig. 15.)



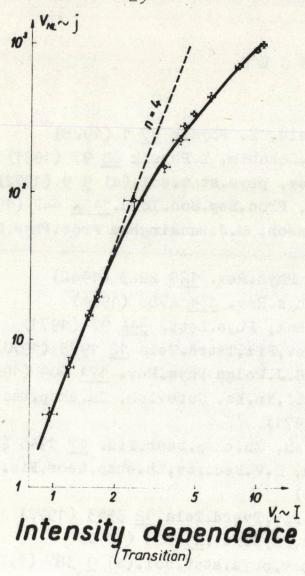
F I G. 15.

Futher interesting information can be obtained from comparison of the total distributions of the photocurrent maxima $p/j_L/$ of the linear detector /i.e. the laser/ and of the $p/j_{NL}/$ function of the nonlinear detector. In the ideal case both $p/j_L/$ and $p/j_{NL}/$ show Poissonian form with $p/j_{NL}/$ "flatter" than the $p/j_L/$ distribution. $p/j_L/$ remains nearly constant as the coherence properties of the laser beam are varied where as $p/j_{NL}/$, which is more sensitive to the coherence, was shown in Budapest to become quickly distorted to an exponential form [37].

In summary, it can be concluded that these experiments verified clearly the existence of the electron emission via the multiquantum photoeffect.

* *

The question now remains, what happens when the light intensity is increased further. On the base of the work done by Keldysh, Bunkin, Fyodorov and Silin we are led to except emission by the tunnel process at extreme high intensities. For the transition range between the pure photoelectric process and the tunnel process, however, existing theory does not describe the explicit form of the intensity dependence, but it can be predicted qualitatively that the increase of the photocurrent should be slower than the j~In power dependence it displays in the lower intensity range. The consequencies of the general theory of Reiss in the case of metals are to lead to similar results. Kovarsky's arguments [3] are similar This prediction has been verified by us in experiments with Au and Ni cathodes and ultrashort high intensity Nd: glass laser pulses [38], when emission is free from Richardson background. (Fig. 16.)



F I G. 16.

The results have demonstrated that the predicted dependence indeed deviates from the Iⁿ form. Control measurements were performed for the intensity dependence of the effect using the green second harmonic of the Nd laser and also for the strong polarisation dependence of the effect. These checks seem to indicate that in this high intensity range the falling order of the nonlinear photoeffect leads to a chage in the character of the electron emission process. However, there is no prospect of us being able more quantitative conclusions until we possess an exact theory, and a more satisfactory knowledge of the structure of ultrashort pulses.

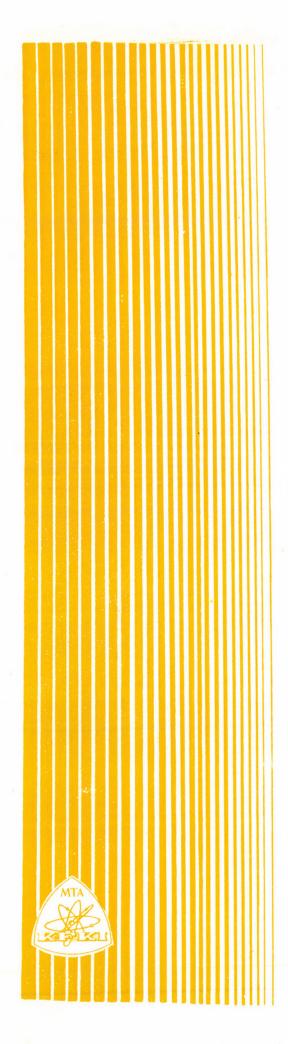
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