Nonlinear photoemission from W and Cu investigated by total-yield correlation measurements

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Nonlinear photoelectric emission from Cu and W is investigated by subpicosecond laser pulses. Using a pump and probe technique, total-yield correlation measurements are obtained without space-charge saturation. The effects of the heating of the conduction electrons induced by the laser pulses are shown, considering the peak to background contrast ratio of the experimental correlation trace. The present analysis clarifies the consequences of a transient nonequilibrium temperature difference between electrons and the lattice on the correlation trace, confirming for W the presence of a thermally assisted photoemission mechanism. [S0163-1829(97)05628-2]

I. INTRODUCTION

Correlation techniques are of widespread use in obtaining information about the temporal dynamics of processes induced by short laser pulses, and, in particular, the temporal evolution of nonlinear electron photoemission from metal surfaces.[1–4] Indeed, the extracted charge produced by a nonlinear emission process, as a function of the delay between spatially overlapped pulses, yields a signal that is a correlation trace of the incident pulses. In a first approximation, depending on the nature of the conduction electrons—i.e., sp electrons (noble, alkaline, and earth alkaline metals), d electrons (transition metals), f electrons (rare-earth metals)—and the energy released by the laser pulse, two different regimes can exist. One regime concerns the nonlinear photoemission via multiphoton absorption, the other arises from a thermally assisted emission process.[5]

Previous experimental investigations have already indicated how to utilize multiphoton yield correlations to study different aspects of laser-solid interaction.[2,3] However, when thermally assisted photoemission starts contributing, the nonlinear response of the photoelectric process is modified, and so are the correlation data.[1]

This paper discusses the effects of the heating of conduction electrons in Cu and W on the correlation trace obtained from nonlinear photoemission processes induced by sub-ps laser pulses, without space-charge saturation. Moreover, to interpret the correlation data, we set forth a model that accounts for thermally assisted photoemission, starting from the theory developed by Bechtel and Bloembergen (B-B model).[5]

The photoelectron signal $S(\tau)$ in a pump and probe experiment with two beams of equal intensity, assuming a pure multiphoton process, and an instantaneous response of the electrons to the applied laser field, is written as

$$S(\tau)\propto\int_{-\infty}^{\infty}[E(t)+E(t-\tau)]^{2n_0}dt, \qquad (1)$$

where n_0 denotes the integer part of $\phi/h\nu+1$ (ϕ is the work function of the material, $h\nu$ is the photon energy) and $E(t)$ is the electric field associated with each pulse. While Eq. (1) is adequate to describe correlation data in multiphoton processes such as those induced on Cu surfaces,[3,6] it is unsuitable for use in interpreting the correlation data obtained from total yield thermally assisted nonlinear photoemission processes such as is observed in W. In fact, it has been experimentally demonstrated[1,6] that the photoemitted charge from

W, using short laser pulses with $h\nu \cong 2$ eV, does not have a dependence on intensity typical of an intrinsic three-photon process, i.e., a slope 3 on a charge versus intensity log-log plot ($\phi \cong 4.3$ eV for W and consequently $n_0 = 3$ at $h\nu \cong 2$ eV). Instead, it is found to have a slope close to 4. For a fourth-order nonlinearity it is expected that the correlation function, on the basis of Eq. (1), has a peak-to-background contrast ratio (PBCR) of 35:1. This theoretical prediction disagrees with the lower PBCR found experimentally.[1] To explain this disagreement space-charge saturation conditions were invoked.

In the present experiment, not affected by space-charge saturation, a lower than expected PBCR in the correlation trace on W is found, and a theoretical estimate that accounts for the PBCR observed experimentally is proposed to interpret nonlinear photoemission on W.

II. EXPERIMENTAL SETUP

The laser system, providing 150-fs pulses at a center wavelength of 600 nm, is described elsewhere.[6] To perform total yield correlation measurements, each pulse is divided into two, and the relative delay is varied by a computer-controlled stepper motor driven delay line with 0.1-μm resolution. Then, the pulses are overlapped noncollinearly onto the sample at nearly normal incidence over an area of about 400 μm^2. The experimental setup allows for the simultaneous detection and comparison of the total yield nonlinear photoemission correlation and the background free second-order autocorrelation obtained from a potassium dihydrogen phosphate crystal. The above features permit obtaining the absolute reference for the zero time delay position unambiguously.

A disk of polycrystalline Cu and a similar sample of W are used as target photocathodes. Sample surfaces are polished according to the procedure reported in Ref. 6. The residual gas pressure in the experimental chamber is in the μPa range. To avoid space-charge problems, a positive 30-kV dc bias voltage was applied to an anode grid mounted parallel to each cathode at a distance of 2 mm. Both cathodes are shorted to ground through a resistor whose voltage drop is used to measure the yield with a calibrated gated integrator. As reported by other authors, the desorbing action of the laser should provide a satisfactory cleaning action on the sample surface.[5]

III. RESULTS

Experimental traces of the total yield autocorrelation measurements from Cu and W are shown in Figs. 1 and 2, respectively. In both cases, the correlation traces are obtained at a low single pulse intensity level in order to avoid space-charge saturation in the wings and in the peak of the correlation. Both correlation traces are normalized with respect to the background mean level, so that it is possible to read directly the PBCR on the vertical scale of the plots. In the inset of each figure is a log-log plot of the extracted charge versus the intensity of a single laser beam. The inset plots are useful in obtaining the order of nonlinearity (slope) and as a test of space-charge saturation (bending at high intensity).

Figure 1 reports measurements obtained from a Cu surface. In this case the log-log slope of the charge versus intensity plot (shown in the inset) and the correlation data are in agreement with a pure three-photon process ($\phi \cong 4.6$ eV for Cu and consequently $n_0 = 3$ at $h\nu \cong 2$ eV). The experimental correlation trace is fitted with the expected third-order correlation function, with a PBCR of 10: it is calculated for a noncollinear beam geometry on the basis of Eq. (1) (with $n_0 = 3$), assuming a single sided exponential laser-pulse profile. The calculated correlation function has only a multiplicative factor as a free fitting parameter.

Figure 2 shows the correlation trace obtained for the W sample. This correlation measurement is not affected by space-charge saturation, but still we have a lower PBCR compared to that expected for a fourth-order nonlinearity. To explain the value of PBCR obtained in this measurement, we

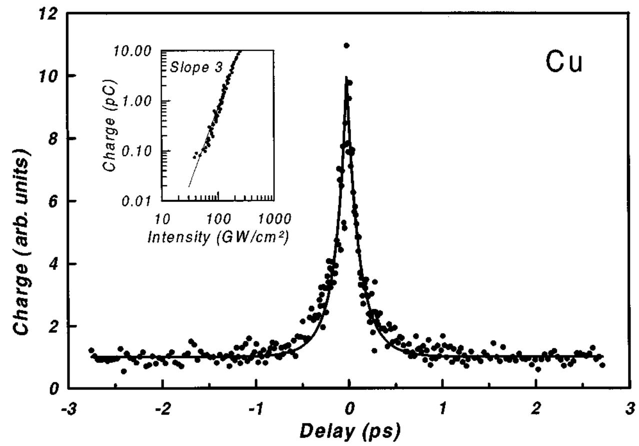

FIG. 1. Total photoemission yield correlation trace obtained for Cu in the absence of space-charge saturation. Single pulse intensity is $\approx$60 GW/cm^2. The fitting curve is superimposed on the experimental correlation trace (see text). The total photoemitted charge vs laser-pulse intensity, reported in the inset, shows a slope close to 3 (indicated by the line) in the overall intensity range. In both measurements, each point represents a mean value over 10 laser pulses.

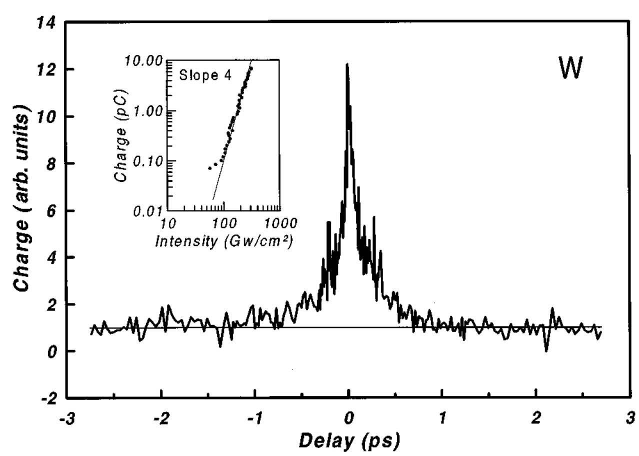

FIG. 2. Total photoemission yield correlation trace obtained for W in the absence of space-charge saturation. Single pulse intensity is $\approx$110 GW/cm^2. The total photoemitted charge vs the laser pulse intensity, reported in the inset, shows a slope close to 4 (indicated by the line) in the overall intensity range. In both measurements, each point represents a mean value over 10 laser pulses.

must consider the contribution of thermally assisted processes.

IV. DISCUSSION

The proposed phenomenological B-B model to explain photoemission from W is based on the assumption that the total photoemitted current density is composed of a weighted sum of nonlinear current densities, where each partial current density is characterized by an integer order nonlinear coefficient n.[5]

For low laser intensity levels, the dependence of the current density on laser intensity given by the B-B model is identical to that deduced by conventional lowest-order perturbation theory, assuming one keeps only the term with $n = n_0$ in the partial current expansion.

For high laser intensity levels, nonequilibrium heating of electrons modifies the photoemission characteristic. Nonequilibrium heating results when the laser-pulse temporal width is shorter than the electron-phonon relaxation time, which is in the range of 1 ps.[1,7] Since in a metal the thermal capacitance of the electron system is much smaller than that of the lattice, a sub-ps laser pulse leads, immediately after the interaction, to high electronic temperatures while leaving a cold lattice. In this way, thermionic effects are avoided, and electrons have sufficient thermal energy to stimulate temperature-induced nonlinear current densities with n *lower* than n_0. Such a temperature-induced regime is referred to as ''thermally assisted photoemission.''[5]

Thermally assisted photoemission can be intuitively explained by considering that the tail of the Fermi function due to thermal excitation is raised substantially in energy (towards the vacuum level) as a consequence of laser excitation. Hence, partial currents with order lower than n_0 will contribute to the total current. Among the induced effects on the experimentally observable photoemission characteristics are (1) a modified nonlinear behavior of the charge versus intensity traces with respect to multiphoton photoemission, (2) distortions of the expected autocorrelation traces of the laser pulses in the extracted charge, and (3) a different relation between the experimentally determined nonlinear order and PBCR.

The photocurrent characteristics of W in the femtosecond high peak intensity regime are explained with a substantial contribution from the highest-order thermally assisted current J_{n_0-1}, considering that it seems unlikely that lower-order thermally assisted currents should give significant contributions.[5] Using the temperature profile calculated from the two temperature model equations (see Ref. 1 and references therein), the charge emitted by each partial current is computed by numerical integration. The model indicates that, in our intensity range, for a W sample with $n_0 = 3$ ($h\nu = 2$ eV, $\phi = 4.3$ eV), the calculated slope for $J_2(r,t)$ is close to 4. Since the measured slope is also close to 4, the total current is assumed equal to $J_2(r,t)$.

On this basis it is possible to estimate the total yield correlation function PBCR for W. The correlation function $S(\tau)$ is given by

$$S(\tau)=\beta\langle[I^2+I_\tau^2+4II_\tau]G(T_e(I+I_\tau))\rangle, \qquad (2)$$

$$G(T_e(I+I_\tau))=T_e(\mathbf{r},t)^2F\left(\frac{2h\nu-\phi}{k_BT_e(\mathbf{r},t)}\right),$$

where k_B is Boltzmann's constant, T_e is the electronic temperature, $\langle\ \rangle$ indicates temporal averages, β is a proportionality factor, and I and I_τ are shorthands for $I(t)$ and $I(t-\tau)$ that indicate the instantaneous intensities associated with each pulse. The function $G(T_e(I+I_\tau))$ depends on the total intensity through the electronic temperature profile, $T_e(r,t)$, determined by the absorbed energy up to time t.[1] In G, only the sum of the intensities of the laser beams and their relative delay are considered in expressing the total intensity, thus ignoring possible interference effects. The intensity correlation term is written neglecting terms whose phase oscillates at frequencies of a multiple of ν, because they are integrated by the detector's slow response; interferometric terms (terms whose phase oscillate with the relative delay τ) are neglected because of the noncollinear beam geometry.

Experimentally it is observed that, with a *single* laser pulse, the relation between extracted current density and laser intensity is

$$J_2=\alpha I_p^{n_e}=\beta\langle I^2G(T_e(I))\rangle, \qquad (3)$$

where I_p is the laser pulse peak intensity [energy/(area $\times$ temporal width)], n_e is the experimentally determined order of nonlinearity, and α is a proportionality factor.

From Eqs. (2) and (3) it is possible to obtain a relation for PBCR, which depends on the measured value of n_e.

For zero time delay ($\tau=0$) the correlation function is

$$S(\tau=0)=6\beta\langle I^2G(T_e(2I))\rangle=\tfrac{6}{4}\beta\langle(2I)^2G(T_e(2I))\rangle$$
$$=\tfrac{3}{2}\alpha(2I_p)^{n_e}, \qquad (4)$$

where the relation that holds for a single pulse has been used.

Considering the case $\tau\to\infty$, we have simply that

$$S(\tau\to\infty)=2\alpha I_p^{n_e}, \qquad (5)$$

since pulses with great relative delay are expected to add their effects. The PBCR is now calculated as

$$\frac{S(\tau=0)}{S(\tau\to\infty)}=\frac{3}{2}\,\frac{\alpha(2I_p)^{n_e}}{2\alpha I_p^{n_e}}=6\times2^{n_e-3}. \qquad (6)$$

With the experimental value $n_e \cong 4$ (Ref. 1 and inset of Fig. 2), we expect that PBCR $\cong 12$, if other disturbing concomitant effects are not present (e.g., space-charge saturation). This value is much less than that expected for a fourth-order dependence, but it is in agreement with correlation measurements on W (Fig. 2). In fact, Eq. (6), in spite of the approximation made, clearly justifies the reduction in PBCR, suggesting the presence of a photoemission process strongly influenced by the electron-gas temperature. Finally, these results confirm the significant role of a thermally assisted two-photon electron emission from W induced by ≈ 2 eV high intensity sub-ps laser pulses, whereas, for Cu thermal effects on photoemission seem to be negligible. These results lead one to speculate on the origin of these differences, which could arise from the different properties of the delocalized *sp* conduction electrons of Cu versus the strongly correlated *d* electrons of W. However, only detailed studies of the *e*-*e* scattering and electron correlation mechanism could properly address this question.

V. CONCLUSIONS

In conclusion, this paper reports a correlation in the extracted charge using a three-photon emission process induced by sub-ps laser pulses on a Cu surface. The experimental data compare well with a calculated third-order correlation function of the laser pulses, and this hints to a nearly instantaneous response of the emitted electrons to the applied field. Further thermal effects are negligible in Cu, at least in the intensity range explored.

On the contrary, similar correlation measurements done on W give a PBCR ratio in agreement with the estimate made using a model that takes into account the transient nonequilibrium heating of the electrons. This result confirms that for W the photoemission induced by sub-ps laser pulses is due to a thermally assisted photoelectric process.

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