Modulation of multiple photon energies by use of surface plasmons

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A form of optical modulation at low pulse rates is reported in the case of surface plasmons excited by 1.55-μm photons in a thin gold foil. Several visible-photon energies are shown to be pulsed by the action of the infrared pulses, the effect being maximized when each visible beam also excites surface plasmons. The infrared surface plasmons are implicated as the primary cause of thermally induced changes in the foil. The thermal effects dissipate in sufficiently small times so that operation up to the kilohertz range in pulse repetition frequency is obtained. Unlike direct photothermal phenomena, no phase change is necessary for the effect to be observed. © 2005 Optical Society of America

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Since their discovery[1] in 1957 surface plasmons (SPs) have been studied in a variety of applications of broad significance.[2] In particular, SPs have long been observed by optical stimulation in thin metal foils in the Kretschmann configuration.[3] In the present study this configuration (Fig. 1) is shown to (1) permit a variety of optical modulations of signals at low frequencies [Fig. 1(a)] and (2) offer a potential means for modulation at high frequencies [Fig. 1(b)]. SPs excited in a thin gold foil can decay by a number of mechanisms related to radiative, acoustic, and thermal coupling. The excitation in the Kretschmann geometry utilizes a transparent substrate such as quartz to increase the wave vector of the incident light k to match the wave vector of the SP (κ) at the given energy in the foil. When the exciting beam is set at the resonance angle θ, computed from the SP dispersion relation, the p-polarization component excites SPs of wave vector $\kappa = nk \sin\theta$, where n is the index of refraction of the substrate. The frequency range for which this occurs depends on the optical properties of the material of the foil and the foil thickness. Simultaneous energy scaling and intensity modulation are reported here for the case of a 1.55-μm communication laser pulsed up to a pulse repetition frequency of 10 KHz. Several unpulsed beams from lasers of visible wavelengths are shown to be pulsed at this same rate by the action of infrared SPs in a thin gold foil, a process henceforth referred to as SP-assisted coupling (SPAC). A method for implementation at much-higher pulse rates that utilizes SP interference is also proposed.

Gold exhibits a well-defined SP in the visible and infrared and thus can strongly absorb incident photons, and the decay can significantly alter the foil at sufficiently high intensities. This is not the same as the local alterations in nonlinear media in more-common cases wherein phenomena such as self-focusing occur. In the present situation the SPs act as an intermediate and essential excitation. Moreover, thermal effects dissipate relatively quickly in a gold foil because of the geometry and the high thermal conductivity. This permits relatively rapid repetition rates for associated processes.

A diagram of the core apparatus and optical paths is shown in Fig. 1, which displays the Kretschmann configuration with additional laser beams incident on the gold foil within the exact same foil area. The infrared pump laser ($\lambda_p = 1550$ nm) is pulsed with a mechanical chopper at lower frequencies and by use of an acousto-optic modulator for higher frequencies. The low-power visible probe laser beams (He–Cd at $\lambda_{pr1} = 442$ nm, Ar^+ at $\lambda_{pr2} = 515$ nm, and a He–Ne at $\lambda_{pr3} = 632.8$ nm used here with typical power levels in the range 1–20 mW) are not pulsed and are initially and independently set so that minimal reflection

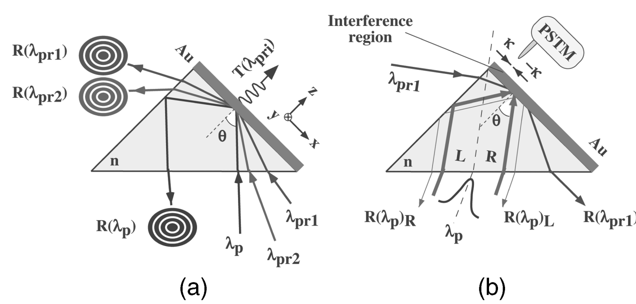

Fig. 1. Schematics of the experimental arrangements for SPAC. The 29.5-nm-thick gold film (xy plane), vacuum evaporated on the side of a right-angled triangular quartz prism (index of refraction of $n = 1.46$), supports plasmon excitation with optimized absorption for the information-encoded beam of wavelength $\lambda_p = 1550$ nm. (a) Simultaneous excitation of the SPs at several visible-photon energies (λ_{pri}, $i = 1, 2, \ldots$) is modulated by the SP excitation due to the higher-power infrared photons (λ_p). This modulation can be detected in all the reflected beams $R(\lambda_{pri})$ ($i = 1, 2, \ldots$) in the case of p polarization and in all the reflected and transmitted beams $T(\lambda_{pri})$ ($i = 1, 2, \ldots$) in the case of s polarization. (b) Left (L) portion of the spatially filtered infrared excitation beam incident at $\theta = 43.6°$ with respect to the film's normal ($\|\hat{z}$) is reflected from the opposite side of the prism back to the film plane, resulting in an overlapping incident region. The corresponding overlapping region at the surfaces of the film will sustain two counterpropagating SPs ($\pm\bar{\kappa}$) that will interfere. The resulting fringes can be imaged with a PSTM. Modulation with SPAC is thus possible in this region.

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occurs. Thus each beam is incident at its SP excitation angle for the beam's wavelength ($\theta_p = 43.6°$, $\theta_{\mathrm{pr}1} = 60°$, $\theta_{\mathrm{pr}2} = 53°$, $\theta_{\mathrm{pr}3} = 46.9°$). When the infrared beam is on at the same time as the visible beams, the SP excitation conditions are modified so that there are measurable reflected photons in the visible. If the infrared beam is off (infrared SPAC disabled) then the reflected visible light is minimal. The reflected beams are observed to be pulsed, and a lock-in amplifier is used to differentiate the pulsed beams from any unpulsed light. The amplitudes of modulation of the probe beams drop continuously to zero for the same modulation frequency if the polarization state of the pump infrared beam is varied continuously from p to s. However, the polarization states of the incident visible beams influence the power levels of only the reflected (modulated) beams. The pulsing action during each cycle of modulation takes on the form of a physical redistribution of the power density of the reflected probe beams similar to a lateral convergence for moderate infrared power levels and a divergence for higher power levels. This is shown in Fig. 2 for $\lambda_{\mathrm{pr}1}$, where such a redistribution is demonstrated in a sequence of spatial beam profiles recorded at increasing infrared power levels. As can be seen, an initial squeezing of the unmodulated reflected $\lambda_{\mathrm{pr}1}$ beam evolves into a laterally distributed beam. Simultaneously, an increase in the power level of the reflected visible beams is also measured that depends on the infrared λ_p power level for moderate ranges. This dependence was measured to be quadratic and well described by the fit $\Delta f(x) = ax^p$, where $p = 2.03 \pm 0.02$, f denotes the reflected $\lambda_{\mathrm{pr}1}$ power, and x stands for the incident λ_p power. The latter, measuring the strength of the SPAC, can be attributed to a perturbation of the resonance conditions of the SPs excited by the visible beams as a result of the infrared excitation. In the experiment depicted in Fig. 1(a) reflected visible beams appear, when observed at long distances from the gold film, to be diffracted as in the pattern of a circular aperture leading to a circular interference pattern. It seems likely that the complicated competition between the SPs excited by the infrared photons and those stimulated by the visible photons within the same region is important on short time scales. On the other hand, operating on longer time scales, the energy deposited into the gold foil by the decaying SPs thermally modifies the local optical properties of the foil. Whereas the former process, which is distinctly different from second-harmonic generation, requires modeling excitation of SPs at minimally two separate energies (ω_p and $\omega_{\mathrm{pr}1}$), the latter essentially requires the temperature dependence of the complex dielectric function of the gold foil (in conjunction with the induced macroscopic variations such as volume expansion of the involved multilayer). For example, a Gaussian modulated volume expansion of the excitation region can result in a variation in the local electron density, which in turn shifts the local value of the dielectric function and thus alters the SP dispersion relation. We therefore surmise that the interference in the various parts of this region gives rise to the formation of the observed rings. A theoretical calculation of the results is in progress. Also noteworthy is that, although it is much weaker (for the used visible/infrared power level ratio, which can also be >1), we observed the reverse process, i.e., the SPAC-mediated stationary effect of the visible $\lambda_{\mathrm{pr}1}$ beam on the infrared λ_p reflected beam to be measurable. The reflected λ_p beam undergoes a similar deformation as a result of turning any of the visible $\lambda_{\mathrm{pr}i}$ ($i = 1, 2, \ldots$) beams on. This interplay is minimal when the polarization state of the $\lambda_{\mathrm{pr}1}$ beam is shifted from p to s and absent when λ_p is also s polarized. Owing to the local variation induced in the dielectric function of the metal film, a shift of the polarization state of the $\lambda_{\mathrm{pr}1}$ beam from p to s results in a transmitted $\lambda_{\mathrm{pr}1}$ beam that exhibits no dependence on the power level of the modulating infrared beam. When modulated, however, this transmitted beam also diffracts into a similar (complementary) ring system. Variation of any of the parameters of the infrared beam, such as angle of incidence, polarization state, intensity, and frequency constitutes a modulation channel for all the visible beams. A distinct advantage of this system is its basic simplicity.

We now return to the diagram in Fig. 1(b), which shows the apparatus for the case in which a standing SP is initiated through the infrared beam region

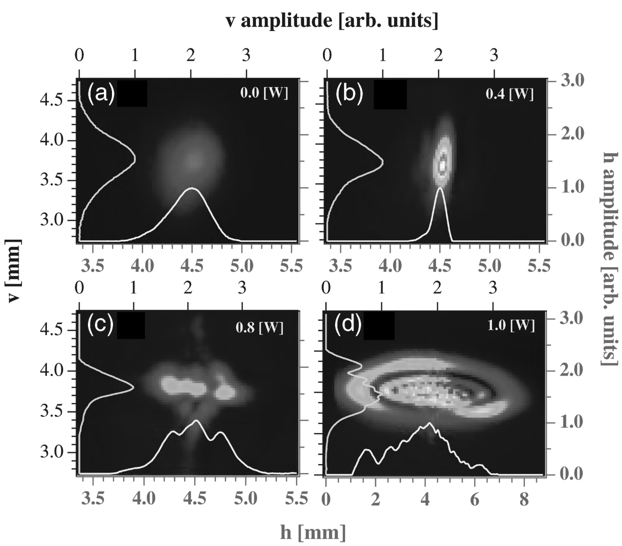

Fig. 2. SP-assisted spatial modulation of the reflected probe beam $R(\lambda_{\mathrm{pr}1} = 442\ \mathrm{nm})$. (a) 20-mW beam loses energy to the SPs in a 29.5-nm-thick gold foil to generate a 0.3 mW of reflected beam, which is recorded a few centimeters (≈8 cm) from the exit face of the prism in the hv plane, a plane perpendicular to the direction of $R(\lambda_{\mathrm{pr}1})$. The resonance conditions are subsequently modified by the excitation of SPs as a result of the infrared beam λ_p, as is evident from the sequence of profiles displayed in (b) and (c), which were recorded in the hv plane as a function of increasing power levels of λ_p as labeled. The vertical and horizontal line profiles are taken from the point of maximum intensity in the images. In (d) the measurement is repeated at a further distance (≈20 cm) from the exit face of the prism. During this process a 44% relative increase of the $R(\lambda_{\mathrm{pr}1})$ beam power is measured, while the horizontal FWHM decreases initially from (a) 0.5 mm to (b) 150 μm, after which it increases to (c) ~1 mm.

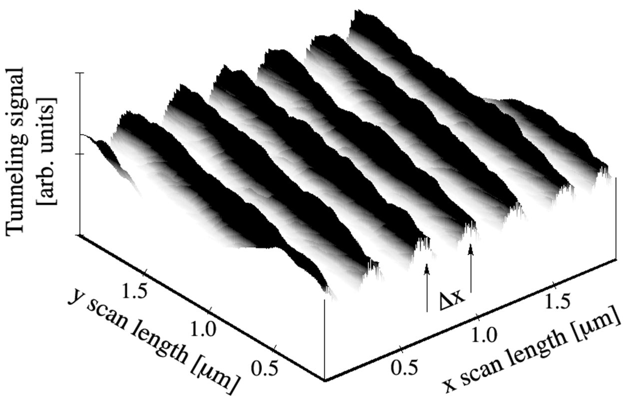

Fig. 3. Proposed implementation of the high-frequency modulation. Standing SPs excited at $\lambda = 632.8$ nm on a 55-nm-thick gold film as probed by the PSTM tip when scanned over a $3\lambda \times 3\lambda$ region. By analyzing the fast Fourier transform of the image, we confirm that no other periodicities are present and obtain a fringe separation of $\Delta x = 302$ nm ($\approx \lambda/2n \sin\theta$, where $\theta = 46°$ is the peak resonance angle). Similar interference can be observed at $\lambda = 1550$ nm on a 30-nm gold film. Modulating the infrared beam can thus accomplish a modulation in the visible probe beams.

on the foil. In this case the spatially filtered beam is expanded and impinges on a 45° prism centered on the apex internally with the gold foil on one face. The critical angle being less than the resonance angle, the portion of the beam (L) that illuminates the bare side of the prism near the apex is totally internally reflected to the foil-bearing face. With the beam set at the resonance angle, the two portions of the beam (L and R) are therefore incident from opposite sides of the foil and engender SPs of opposing wave vector. From our observations it is noted that a standing SP can be engendered during the time of a pulse and that a pulse of sufficiently short interval can be used that is much shorter than the time needed for thermal transfer across the period of the standing wave. In this case the interval between pulses can be set to allow dissipation of the pattern before any subsequent pulse arrives. We estimate that, if heat is suddenly liberated and maintained in the mode proportional to $\cos^2(x/\lambda)$ in a medium with thermal diffusivity K, then the time characterizing the decay of the space-varying temperature distribution toward a constant value is $\lambda^2/4K$. In a gold foil with $K = 1.17\ \mathrm{cm}^2\ \mathrm{s}^{-1}$ and for $\lambda = \lambda_p = 1550$ nm, the characteristic time is ≈ 5 ns. As such, the rapid transfer of an information train of pulses may be possible and indeed is not possible at lower pulse rates. Since the standing-wave pattern affects any additional visible beam of light, a second form of optical modulation can be enabled at much-higher pulse rates, albeit only at the higher rates. Observation of such a grating is reported here by the use of a photon scanning tunneling microscope (PSTM)[4] and is shown in Fig. 3, in which a standing longitudinal wave of the collective electronic surface charge distribution on the two foil surfaces[5] is imaged. This provides a periodic distribution of the thermal effects introduced by the phonon–SP coupling. After the stimulating beam is turned off, the high thermal conductivity of gold removes the periodicity in the time it takes the heat transfer to travel a fraction of the grating wavelength. This time interval then determines the information transfer rate to a visible-light beam that impinges on the grating. This effect remains to be demonstrated, but the concept is described so that further work might be stimulated for high-data-rate applications.

Modulation of visible light by an infrared communication laser has been demonstrated within the audio range of frequencies. Audio signals carried by infrared light can therefore be converted into a range of colors. Similarly, red, green, and blue laser wavelengths can be modulated by infrared signals that carry visual display information since visual persistence allows use of low-frequency modulation. Other color combinations can be used as necessary to create a full-color display from separately modulated infrared signals. Both concepts presented are subject to caveats in any potential applications. A number of parameters remain to be determined, including the temperature of dependence of the effects. However, the basic effects can be expressed in a straightforward manner in a relatively simple experiment.

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