Light Emission from Inelastic Electron Tunneling

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We report the discovery of a new method for the generation of light. This technique yields a broad-band ligh source with a high frequency linear cutoff which is dependent only upon the applied voltage through the quantum relation $h\nu_{co}=|eV|$. The light source consists of a metal-insulator-metal tunneling junction. The effect can be interpreted in terms of inelastic tunneling excitation of optically coupled surface plasmon modes.

This Letter reports the discovery of a new method for the generation of light. This technique yields a broad-band light source with a high-frequency linear cutoff which is dependent only upon the applied voltage through the quantum relation $h\nu_{co}=|eV|$. The light source consists of a metal-insulator-metal tunneling junction. Light is observed visually to be emitted uniformly over the junction area regardless of the polarity of the applied voltage. The quantum cutoff condition, $h\nu_{co}=|eV|$, can be deduced from conventional optical spectra as well as from a new spectroscopic technique embodying this device. The effect can be interpreted in terms of inelastic tunneling[1-5] excitation of optically coupled surface plasmon modes present in the tunneling junction. For succinctness we term the effect "light emission via inelastic tunneling" (LEIT). We note that other workers have observed light emitted from metal-insulator-metal structures.[6,7] In these cases the oxides were much thicker (300 to 500 Å as opposed to ~30 Å in our case). Light was observed from various spots on such structures and it was believed that other electron transfer mechanisms, such as carrier injection, were involved. The linear quantum cutoff was not observed in such cases since significant inelastic tunneling would not be possible through such thick oxides.

The tunneling junctions are of the type used in elastic tunneling studies of standing waves.[8] That is, a 500-Å aluminum film was oxidized and then crossed with a counter electrode. The counter electrodes were either Ag, Au, Pb, or In of 200 to 300 Å thickness, evaporated with the substrate at 77°K. In addition, a mild etch was used to roughen the counter electrode and to render it slightly porous. This was done to provide optical output coupling to surface plasmon modes in the junction. The junctions were then heated in air at 150°C to decrease the tunneling conductance and thus limit the current at the peak bias voltage of 4 V. The junction area was 5×10^{-2} cm^2 and had a zero bias conductance of about 10^{-2} mho/cm^2.

When voltage was applied to such junctions, visible light was seen to emanate from the entire junction area. One could observe the emission color change from deep red at low voltage to orange to blue white as the voltage was increased. This showed the effect of the change in ν_{co} in a very striking way. The increase in voltage brings in more and more of the visible spectrum. Typical spectra of Ag and Au junctions are shown in Fig. 1. These spectra were taken with the junctions at 77°K. The effect can be observed at room temperature, but high voltage stability was better at 77°K. The change in ν_{co} with voltage is seen very clearly. The linear decrease in intensity as ν_{co} is approached is also evident. We will not discuss the detailed form of the broad spectra at present. For voltages exceeding the bulk plasma frequency in Ag we observe a small peak due to bulk plasma radiation in addition to the large broad-band spectrum. This bulk emission peak has recently been shown to be due to electron injection into Ag.[9] The broad-band emission is not significantly different when the polarity of the voltage is reversed. The relation $h\nu_{co}=|eV|$ is, of course, unaffected. When the direction of the

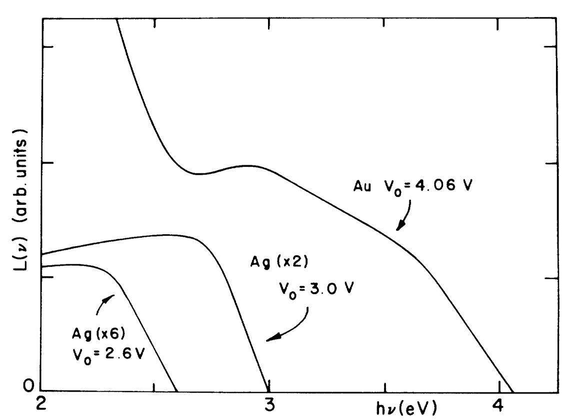

FIG. 1. A plot of $L(\nu)$, the photon flux per unit frequency bandwidth versus $h\nu$ for an Au and an Ag junction at different junction bias voltages. The junction temperature was 77°K.

tunneling current is reversed there is a change in the intensity, but this is not surprising as the tunneling current is not a symmetrical function of voltage. The light emission was found to be diffuse and essentially unpolarized. The external quantum efficiency (ratio of total photon flux to tunneling current) of this light source was estimated to be about 10^{-5} using manufacturer's photomultiplier data. Optical feedback probably could be used to improve the efficiency in a narrow-band configuration.

In order to gain a deeper insight into the nature of the quantum cutoff condition a different kind of spectroscopy was devised. In this case one uses a narrow band, fixed optical filter with a transmission function $\Gamma(\nu)$ centered around a frequency ν_0. This filter is interposed between the emitting junction and a photomultiplier. The photomultiplier current I_{pm} and its second derivative d^2I_{pm}/dV^2 are measured as the voltage V on the tunnel junction is varied. The results are shown in Fig. 2 and show that I_{pm} "turns on" as V passes through the condition $|V| = h\nu_0/e$ and then I_{pm} increases linearly with voltage. d^2I_{pm}/dV^2 shows a peak when $|eV| = h\nu_0$. $\Gamma(\nu)$ is also plotted in Fig. 2 for comparison and it is clear that d^2I_{pm}/dV^2 as a function of voltage maps out the transmission function. The second derivative curve is somewhat broader than $\Gamma(\nu)$. We believe that this is due at least partially to temperature broadening but this has not been established.

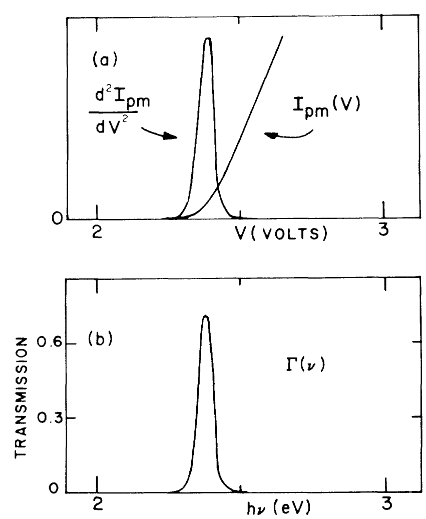

FIG. 2. (a) The photomultiplier current I_{pm} and its second derivative with respect to voltage plotted versus junction voltage for an Ag junction. These quantities are plotted in arbitrary units. The junction temperature was 77°K. (b) The transmittance function $\Gamma(\nu)$ plotted versus $h\nu$.

The results of these measurements can be summarized in terms of a simple model based on inelastic tunneling. An electron tunneling from one metal to another can excite an optically coupled surface plasmon mode in the junction with frequency ν provided that $|eV| \geq h\nu$. This notion of a threshold has been discussed extensively in the case of inelastic tunneling vibrational spectra.[1-3] At very low temperatures the form of the inelastic excitation will be

$$L(\nu) = P(\nu, V)(|V| - h\nu/e)\theta(|V| - h\nu/e),$$

where $L(\nu)$ is the number of photons emitted with frequency ν within an interval $d\nu$. $P(\nu, V)$ is a slowly varying function of frequency and voltage involving the density of surface modes and the inelastic excitation and radiation probabilities. $\theta(|V| - h\nu/e)$ is the step function that reflects the cutoff of photon emission at the quantum condition. Thus, the model predicts a linear cutoff in the spectra as $h\nu$ approaches $|eV|$.

In the transmission filter experiment the measured photomultiplier current will be a product of $L(\nu)$, the quantum efficiency and gain of the photomultiplier. Over the very narrow bandwidth of the optical filter $\Gamma(\nu)$, the photomultiplier factor as well as $P(\nu, V)$ can be considered constants. Thus,

$$I_{pm}(V) \propto \int_0^\infty (|V| - h\nu/e)\theta(|V| - h\nu/e)\Gamma(\nu)d\nu.$$

Taking the second derivative of I_{pm} with respect to voltage gives

$$d^2I_{pm}/dV^2 \propto \Gamma(|eV|/h).$$

The peak in the second derivative of the photomultiplier current is reminiscent of those observed in tunnel current derivatives when molecular vibrations are excited in inelastic tunneling. In that case the frequency was determined by the material in the tunnel junction. This discrete vibration mode excitation led to peaks in the second derivatives of the tunneling current. In the present situation the optical filter "selects" optically coupled surface plasmon modes out of a quasi-continuum whose excitation is to be observed externally.

The technique of measuring the second derivative of the photodetector response yields a new kind of spectroscopy since it can be used to measure the transmission characteristics of an optical filter by simply scanning the voltage. We expect this spectroscopy to have the same temperature dependent resolution as in inelastic electron tunneling spectroscopy.[3] That is, an infinitely sharp transmission function should have a width of $5.4kT$. Thus, the "slit width" of such spectroscopy is expected to be temperature dependent. This effect arises from the thermal smearing of the Fermi distributions in the metal electrodes. Superconducting electrodes should improve the resolution.[3] We have not as yet made a good test of these effects in the light emission case. Similarly, a precise test of the relation $h\nu_{co} = |eV|$ is best carried out at very low temperatures.

In the present Letter, no attempt has been made to evaluate $P(\nu, V)$ which reflects the overall spectral form and the intensity of the light emission. This quantity will depend both on the optical properties of the electrode metals as well as output coupling mechanisms of the radiation from the junction region. We believe that coupling may occur by inelastic excitation of surface plasmon "junction modes."[10-12] Surface plasmon modes on metal surfaces are, of course, well known.[13] Such modes are nonradiative due to momentum nonconservation between the free photon field and the surface mode. It is also established that roughness can bring about optical coupling so that the modes can be made radiative.[13] When two metal surfaces are in close proximity these surface modes interact and form a "slow wave structure." Electromagnetic waves can propagate in the interface region. There then exists a quasicontinuum of modes up to the highest surface plasmon frequency of the metals comprising the junction.[11,12] Again, these modes are expected to be nonradiative in perfectly smooth, flat films, but roughness or other irregularities would cause such modes to radiate. The coupling of tunneling electrons directly to the photon field has been discussed in connection with p-n junctions and was termed "photon assisted tunneling."[4] In that case the junction and surface modes play no role. These aspects of the LEIT effect require further experimental and theoretical studies.

In summary, the LEIT device promises to be a useful broad-band light source whose frequency is limited only by the voltage which the device can withstand. It also promises to form a basic component in transmission or reflection spectroscopy when used in a frequency modulation mode since it incorporates both source and monochromator function in a single electrically scanned unit.

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