Double-Photon Photoelectric Emission from Alkali Antimonides

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Nonlinear response in photoelectron emission from alkali antimonides, such as Cs_3Sb and K_3Sb, under intense laser irradiation was observed directly. The photocurrent was proportional to the incident laser power at lower intensity, but it showed quadratic response at higher intensity. Energy distribution of the emitted electrons in the quadratic region coincided for the most part with that excited by the natural light with a wavelength half that of the laser. These results seem to present experimental evidence for a double-photon absorption process in the nonlinear photoemission caused by intense irradiation.

When high-power laser light falls upon a solid, nonlinear photoelectric emission is to be expected. Theories[1] for double-photon absorption in metal have predicted that the photocurrent will be proportional to the square of the incident power, and this relation was verified experimentally for sodium[2] and gold.[3] For semiconductors the same relation was previously observed in Cs_3Sb,[4] though theoretical work has not yet been published.[5] Thus the nonlinear photoelectric emission under intense laser irradiation has become a rather common phenomenon.

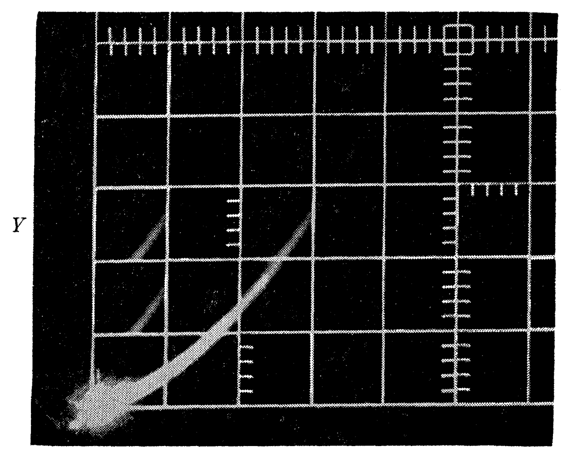


Fig. 1. An oscilloscope trace showing nonlinear photoemission. X and Y coordinates are laser intensity and photoelectric current, respectively. The curve is composed of a large number of spike signals.

In this note, an experimental verification for two-photon process concerning the nonlinear photoemission will be presented by observation of energy distribution of the emitted electrons. The samples used were alkali antimonides such as Cs_3Sb and K_3Sb set in a photomultiplier[6] specially made for the present work. A grid (75 mesh) was inserted substitutionally in the usual position of the cathode, and the sample, prepared on a metal plate, was placed behind the grid at the distance of 1 mm. The light source adopted was a Nd-glass laser (1.06 μ, 1.17 eV) for Cs_3Sb and a ruby laser (0.69 μ, 1.78 eV) for K_3Sb in normal operation. The light beam from the laser head was polarized initially and then

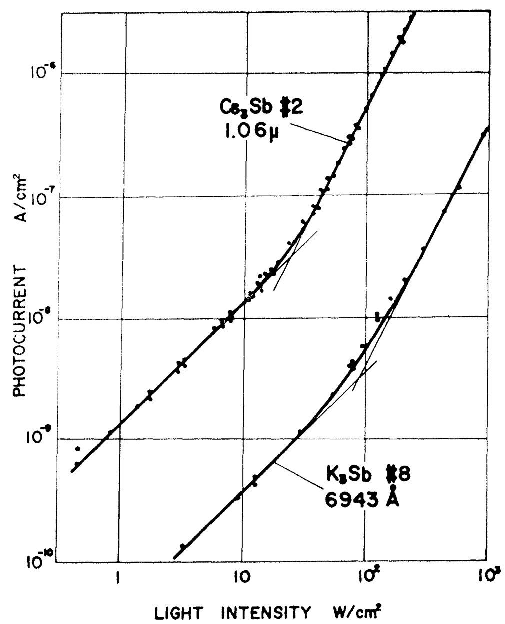


Fig. 2. Plot of photoelectric current versus laser intensity for Cs_3Sb and K_3Sb.

split into two components; one of them was sent into a monitor for intensity (RCA 7102 photomultiplier) and the other was incident on the sample. The signals from both photomultipliers were displayed on an X-Y oscilloscope. Figure 1 illustrates a typical trace showing the nonlinear response directly and clearly. By attenuating the light intensity to appropriate magnitude, the relation between the photocurrent and the light

[1] R. L. Smith, Phys. Rev. **128**, 2225 (1962); I. Adawi, *ibid.* **134**, A788 (1964); P. Bloch, J. Appl. Phys. **35**, 2052 (1964).
[2] M. C. Teich, J. M. Schroeer, and G. J. Wolga, Phys. Rev. Letters **13**, 611 (1964).
[3] E. M. Logothetis and P. L. Hartman, Phys. Rev. Letters **18**, 581 (1967).
[4] H. Sonnenberg, H. Heffner, and W. Spicer, Appl. Phys. Letters **5**, 95 (1964).
[5] Only a final formula of S. Jha's calculation has been cited in Ref. 4.
[6] Made by Hamamatsu TV Co., Ltd., Hamamatsu City, Japan.

intensity is obtainable in a wide range. Figure 2 shows the resultant plot for Cs_3Sb and K_3Sb, though the values of light intensity have some uncertainties because of sectional nonuniform distribution of photon density in the light beam.

The plot shows that the curve consists of two parts, linear and quadratic characteristics. When the laser intensity was increased above the values noted here, the X-Y display disappeared, presumably because of thermionic emission resulting from heating of the sample. By measuring the absolute sensitivity to natural light at the laser wavelength, it was ascertained that the linear part was caused by the long tail of the emission spectrum due to one-photon absorption. Therefore, the location of the shift from linear to quadratic dependence varies with sample preparation.

As a next step, the energy distribution of the emitted electrons was measured by the retarding-potential method. The principle of this procedure is as follows. If the electrons in the quadratic region are emitted by simultaneous absorption of two photons, then the kinetic energy of the emitted electron may be expected to be the same as that due to the absorption of one photon with twice the energy of the laser photons, as is

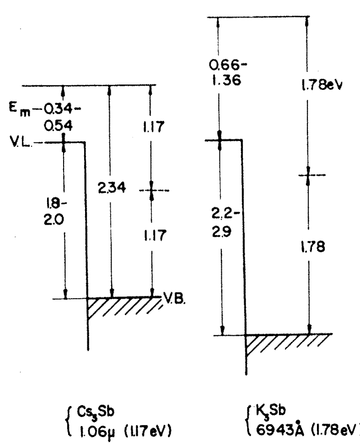


FIG. 3. Schematic energy diagrams for Cs_3Sb and K_3Sb, showing double-photon photoemission. E_m denotes the maximum kinetic energy of emitted electrons referred to the vacuum level (V.L.).

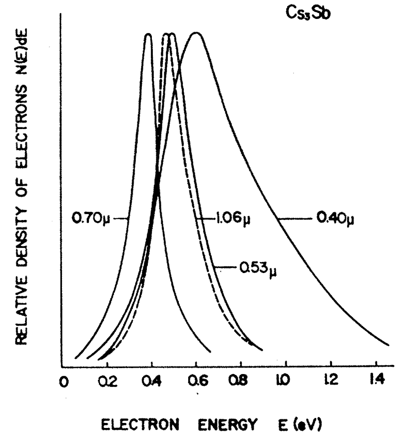


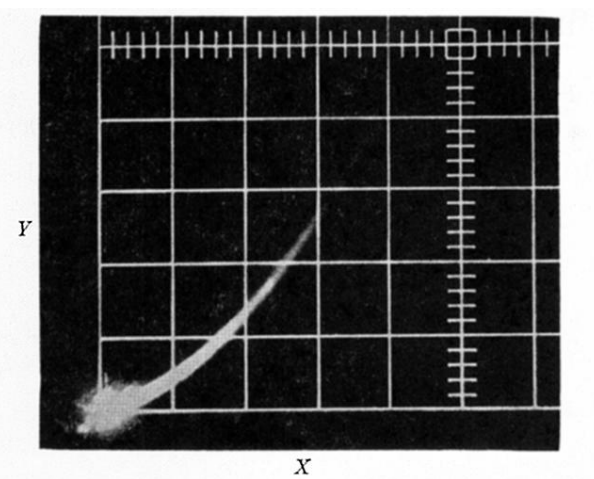
FIG. 4. Normalized energy distribution of photoelectrons from Cs_3Sb under irradiation with natural (solid line) and intense laser (dashed line) light.

easily understood from Fig. 3, based on the energy-band model.[7] The retarding potential was applied between the grid and the sample. Photoelectrons passing through the grid-mesh were multiplied by dynodes of nine stages. It was confirmed, prior to measurement, that the energy distribution of the electrons was not changed significantly by multiplication in the dynodes. As the retarding potential was varied stepwise, the photocurrent at constant light intensity in the quadratic region was measured from an X-Y trace. The energy distribution can be obtained by differentiating the curve of photocurrent versus retarding potential.

Figure 4 illustrates normalized energy distribution curves for Cs_3Sb when it is irradiated by natural and by laser light. From the figure it is clearly shown that energy distribution as irradiated by laser light is similar to that irradiated by natural light with half the wavelength. The magnitude of the electron energy agrees fairly well with that estimated from the model, as shown in Fig. 3.

The results mentioned above seem to present experimental evidence for double-photon photoelectric emission.

[7] W. E. Spicer, Phys. Rev. **112,** 114 (1958); S. Imamura, J. Phys. Soc. Japan **14,** 1497 (1959).



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