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RESONANT MULTIPHOTON PROCESSES IN LASER-INDUCED DESORPTION

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Abstract

The dye laser induced emission of positive ions from the surface of cleaved highly transparent bariumfluoride shows a pronounced dependence on the exciting laser wavelength. Qualitatively, the same spectra are observed for the emission of electrons. For fixed wavelengths, the emission yields of electrons and positive ions follow higher powers of the laser intensity. Consequently, photoemission is interpreted by resonantly enhanced two-photon (for less than ~1.7 MW/cm² laser intensity) respectively five-photon (for higher laser intensity) absorption. Ion desorption then follows from the combination of two holes at a fluorine site.

Even though the bandgap of transparent materials, e.g. ionic crystals, is of the order of 10 eV, visible radiation can be absorbed in such material via multiphoton processes. The effect is of special interest at the surface of these crystals where it can lead to the emission of electrons (photoemission) and surface constituents (desorption). In many cases, laser induced desorption has been attributed to thermal effects /1/. In the present contribution, however, we provide evidence that non-thermal photoemission and the desorption of positive ions from bariumfluoride are closely related and are a consequence of resonantly enhanced multiphoton absorption.

When we irradiate the in vacuo cleaved (111) surface of BaF₂ under ultra-high vacuum with green dye laser light at an intensity of less than 10 MW/cm², i.e. about two orders below the damage threshold /2/, we observe the emission of positive ions from the surface /3/. The analysis of these ions by means of a quadrupole mass filter shows, that they are F⁺, Ba⁺ and (BaF)⁺ (cf. Fig. 1).

Figure 1. Laser-pulse correlated emission of positive ions from BaF₂(111).
(Laser at 532 nm, 10 MW/cm²).

These ions are emitted simultaneously, as can be seen from a first time-of-flight spectrum (Fig. 2).

Figure 2. Time-of-flight spectrum of positive ions, emitted from BaF₂ (111).
(Laser at 532 nm, 3 MW/cm²; distance 3 cm, accelerating potential -30 V).
We also observe the emission of electrons from the surface. From the analysis of the dependence of the electron and ion yields on the laser intensity /3/, we know that both are the result of multiphoton absorption. When we scan the laser wavelength, we observe a pronounced spectral dependence of the yields (Fig. 3) which is practically identical for ions and electrons. It exhibits, however, a dramatic influence of the azimuthal orientation of the surface with respect to the laser polarization. The intermediate resonances remain in position independent of whether two- or five-photon ionization takes place (cf. Fig. 4). Hence, we conclude that the resonance condition

**Figure 3.** Yields for positive ions and electrons from BaF₂ as a function of laser wavelength (5 MW/cm²). In a), b) the projection of the electric field vector \( \vec{e} \), is parallel to the [112] direction, in c), d) \( \vec{e} \), is parallel to [110].

**Figure 4.** Spectral dependence of the electron yield from BaF₂ (111) for different laser intensities:

a) 3.2 x 10⁶ W/cm²
b) 2.8 x 10⁶ W/cm²
c) 2.2 x 10⁶ W/cm²
d) 1.7 x 10⁶ W/cm²
is imposed on the last two photons in each case, in good agreement with the calculated density of states in the bandgap of a non-stoichiometric surface. Together with the observation of the two-photon ionization as such, which requires implicitly occupied states in the middle of the bandgap, this justifies the assumption of the non-stoichiometric composition of the surface. Apparently, at very low laser intensities, which are always present in the wings of the temporal pulse shape, kind of such a preparation can take place, as can be seen from the accidentally measured development of the spectral dependence at one specific surface site (Fig. 5).

Figure 5. Accidentally observed "preparation" with increasing intensity (a) 1 MW/cm², (b) 2 MW/cm², (c) 5 MW/cm²) of one individual spot at the surface. In subsequent measurements at lower intensities, the structure of c) persisted.

The apparent close relationship between electron and ion spectra makes us assume that resonant multiphoton ionization is the origin for the desorption. This idea is supported by the result, that the intensity dependence of the ion yield is the square of that for electrons, indicating that two electrons must be emitted from relatively neighbouring sites to create one positive ion /3/.

From the fact that ions and electrons appear quasi instantaneously at each laser pulse and cease being emitted after the end of each pulse, together with all the above observations, we deduce that our observed phenomena are indeed purely electronic transitions and not related to thermal energy dissipation.

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References
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