Optical, Mass, and Auger Spectra from e-Bombarded KBr*

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Abstract

We have measured the mass spectrum and optical emission lines of neutral potassium atoms ejected from KBr at T = 300°K and 443°K bombarded by 2-keV electrons. The room-temperature data may be complicated by the non-stoichiometry of the alkali-enriched sample surface and seem difficult to interpret. The high-temperature sample, which maintains the proper stoichiometry, produces data in support of gas-phase excitation of alkali atoms desorbed from the surface.

*Sponsored by the Office of Health and Environmental Research, U.S. Department of Energy, under contract DE-AC05-84OR21400 with Martin Marietta Energy Systems, Inc.

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Introduction

Sputtering of metals and semiconductors is generally well described by Sigmund's nuclear-collision cascade theory.[1] In contrast, for ionic solids such as alkali halides,[2,3] sputtering is associated with electronic processes. Investigations on ionic solids have been limited primarily to ground-state species ejected from the sample surface. In recent years, optical emission from bombarded solids has attracted much interest,[4,5] since useful information can be obtained concerning excited-state species produced in the sputtering or desorption process. However, only a few studies have been made, and a need exists for data which will aid in the understanding of excited-state species formed in the sputtering or desorption of ionic crystals. Here, we report optical emission spectra from electron-bombarded KBr, which were obtained simultaneously with measurements of Auger and mass spectra.

Experiment

The experiments were performed in a conventional Auger chamber with pressure in the 10^{-8} Torr range. A crystal of KBr was introduced into the chamber within an hour after cleavage in air. The surface was monitored for contaminants by using a cylindrical mirror Auger analyzer. No detectable amounts of carbon, nitrogen, or oxygen were found. The intensities of the LMM Auger line of potassium and MNN Auger line of bromine were used to monitor the number of electrons injected into the sample for use in determining the beam current dependence of the optical emission intensities. The optical emission was collected with a quartz lens through a MgF_2 window and imaged on the entrance slit of a 0.3-m McPherson spectrometer. The spectral band width was about 0.3 nm. The emission spectrum reported here has been corrected for spectral sensitivity of the detecting system by using an NBS, tungsten-ribbon, standard lamp. The mass spectra were measured with a VG-Spurvac quadrupole mass spectrometer, whose ionization filament was switched off during the optical measurements.

Results

Figure 1 shows the optical emission spectrum of room-temperature KBr at 80° from the sample normal bombarded by 2-keV electrons with an electron flux of 15 $\mu A/mm^2$. The electrons were incident at 10° from the sample normal. The spectrum is composed of sharp lines and a broad band. The broad band centered ~470 nm is of bulk origin[6] and is not of concern in the present context. The wavelengths of the sharp lines coincide with those of the electronic transition from isolated neutral potassium atoms, within the spectral resolution of 0.3 nm.

Figures 2 and 3 show the dependence of the optical line intensities from room-temperature KBr on the beam current. These were measured simultaneously with the Auger or mass intensities to avoid the ambiguities about the number of electrons injected into the samples, the decomposition of the samples, and the number of ground-state potassium atoms. As seen in Fig. 2, the intensities of the optical lines at 404.6 and 766.5 nm increase linearly with increasing beam current in the low current (< 13 μA) region, I, and are proportional to the third power of beam current in the intermediate region, II. In the high current (> 25 μA) region, III, they increase with greater than third power of beam current. On the other hand, the intensities of the Auger lines increase linearly in regions I and II and then deviate from linear dependence in region III. After a long irradiation in region III, the Auger intensities became very weak, indicating decomposition of the sample surface.

As seen in Fig. 3, the number of potassium atoms and the optical line intensities increase linearly with beam current in region I. The number of ground-state potassium atoms increase nonlinearly in region II; while in region III, a saturation seems to be reached. The latter result indicates that the alkali overlayer controls the sputtering rate, with the ground-state alkali atoms evaporating thermally in region III. We believe that the dominant parts of the production of excited-state alkali atoms in regions II and III are associated with electronic processes, although thermal energy is, in part, included in the processes. The nonlinearity of the production rate of the excited-state atoms may be due to secondary processes such as the interactions of the

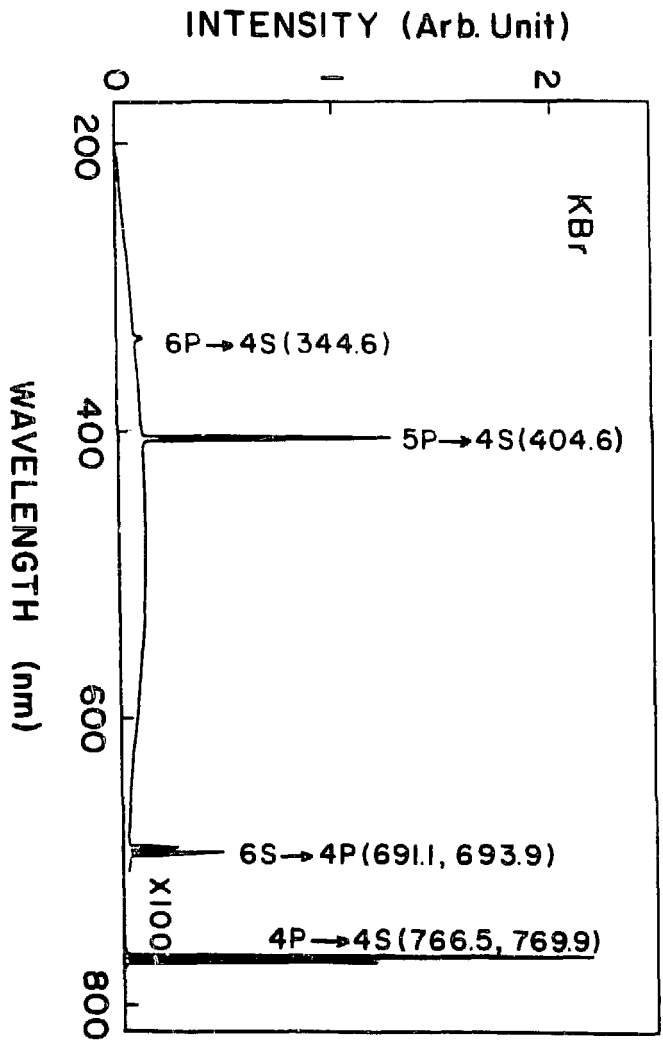

Fig. 1. Optical emission spectrum of room-temperature KBr crystal bombarded by 2-keV electrons with electron flux of 15 $\mu A/mm^2$. The spectral resolution is 0.3 nm.

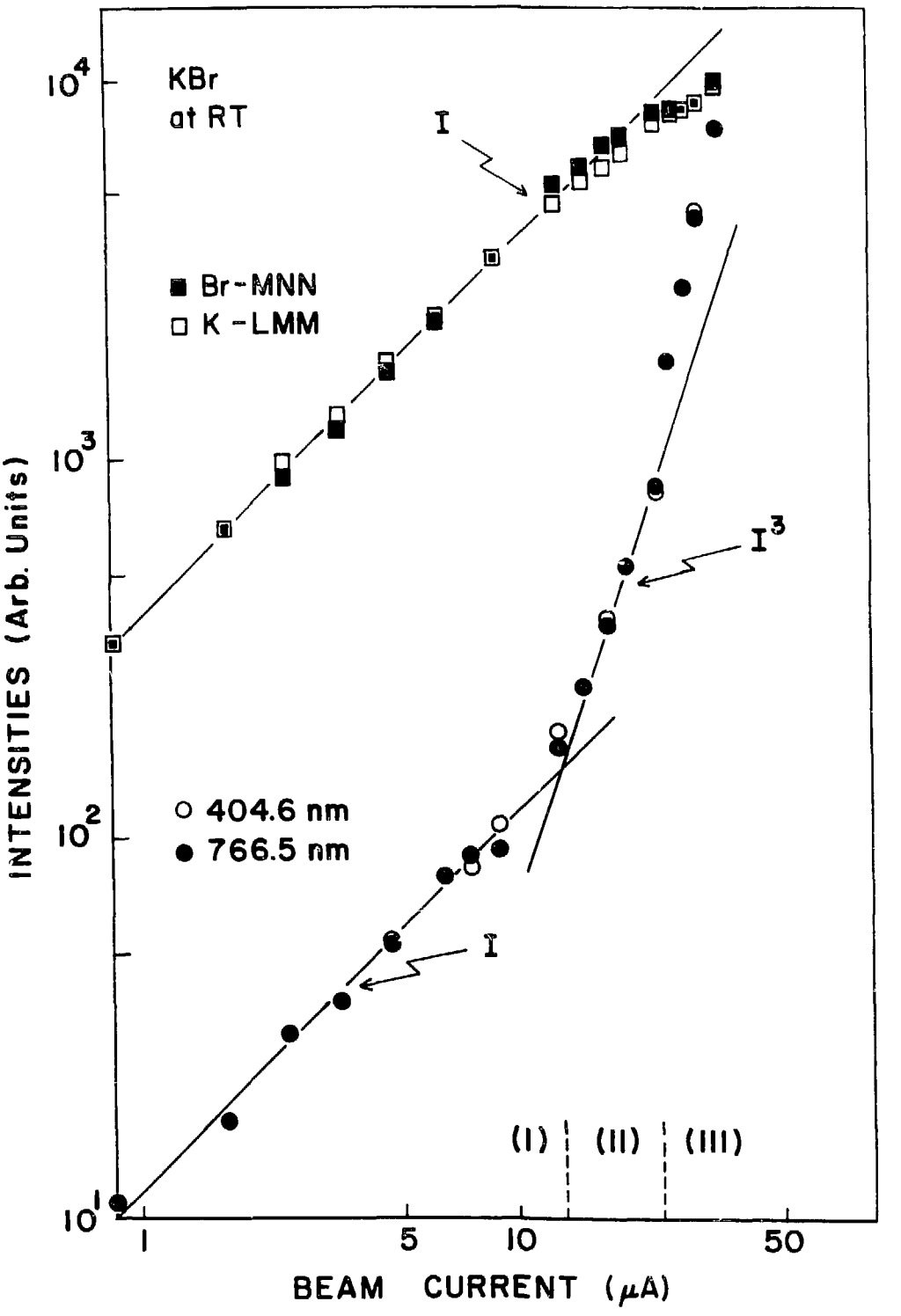

Fig. 2. Beam current dependence of the optical and Auger intensities from KBr at room temperature.

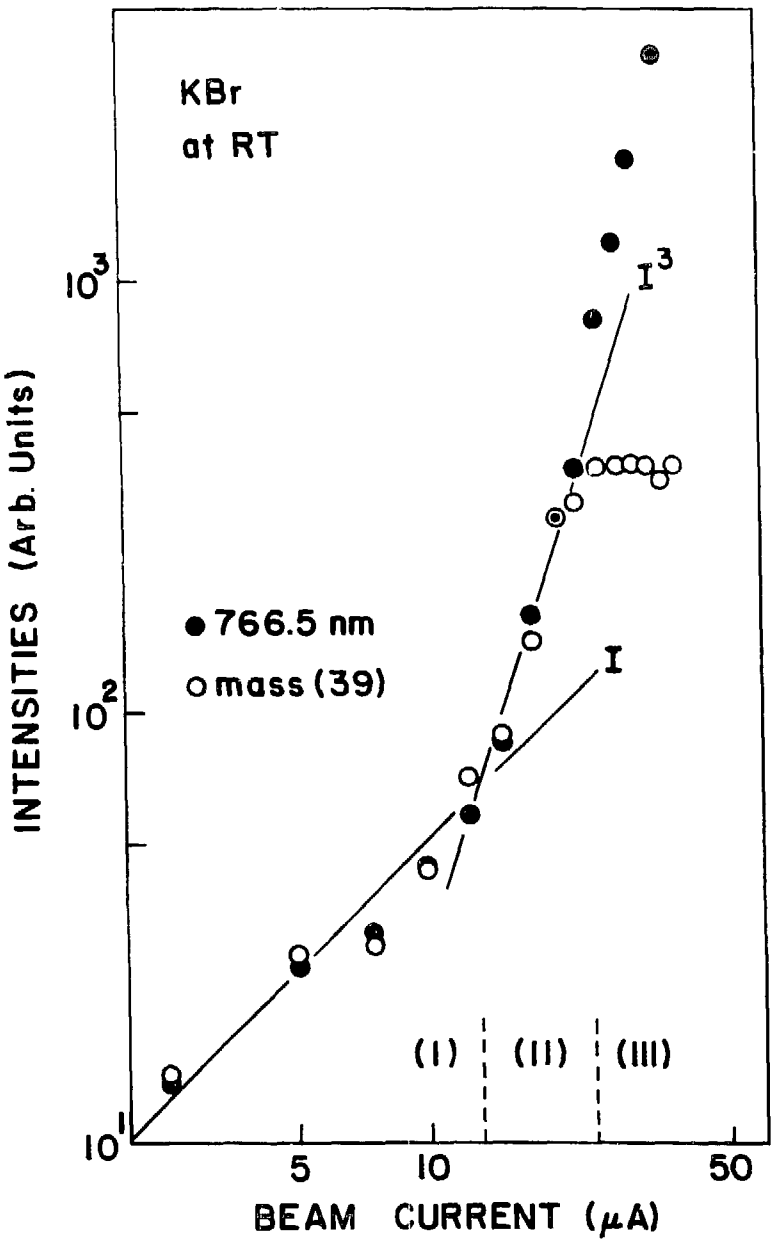

Fig. 3. Beam current dependence of the ground-state and the excited-state potassium atom intensities from KBr at room temperature.

excited-state atoms, ions, and molecules with ground-state atoms, defects, and local vibrations.

In contrast to the complicated room-temperature results, the beam current dependences of the optical line intensity and the number of potassium atoms for KBr heated to 443°K become much simpler. As shown in Fig. 4, the intensities of the ground-state potassium atoms increase almost linearly ($\propto I^{0.88}$), while those of the excited-state potassium atoms increase as $I^{1.65}$ with increasing beam currents.

Discussion

According to existing models of electron sputtering of alkali halides,[8,9] the energy of the incident electrons is lost by excitation or ionization of the halogen sublattice. This energy can be converted into kinetic energy of an H center (a molecular halogen ion occupying one lattice site) which can diffuse to the surface and result in the ejection of halogen atoms. The excess alkali atoms remaining on the sample surface can then vaporize thermally.

Our present results for KBr at T = 443°K are in accord with the results of Walkup et al.,[10,11] who measured the mass distribution and yield of both positive ions and neutral species from electron-bombarded NaCl. Since the surfaces of alkali halide crystals at low temperatures can become alkali-enriched by high-current electron bombardment, they chose to minimize this effect by raising the sample temperature to 570°K. For NaCl, it has been shown that at sufficiently high temperatures (T > 570°K) the surface remains stoichiometric even for high electron beam currents. Under these conditions, they found the yields of Na, Cl, Na_2, and Cl_2 to be linear in beam current from 0.1–10 μA, and the yields of positive ions of these species to be proportional to the square of the electron beam current. These results suggest that the positive ions are produced by gas-phase ionization of neutral atoms and molecules.

Postawa et al.[12] measured the optical emission spectra from NaCl, CsCl and CsI bombarded by electrons or ions as a function of target temperature and distance from the sample surface and concluded that in

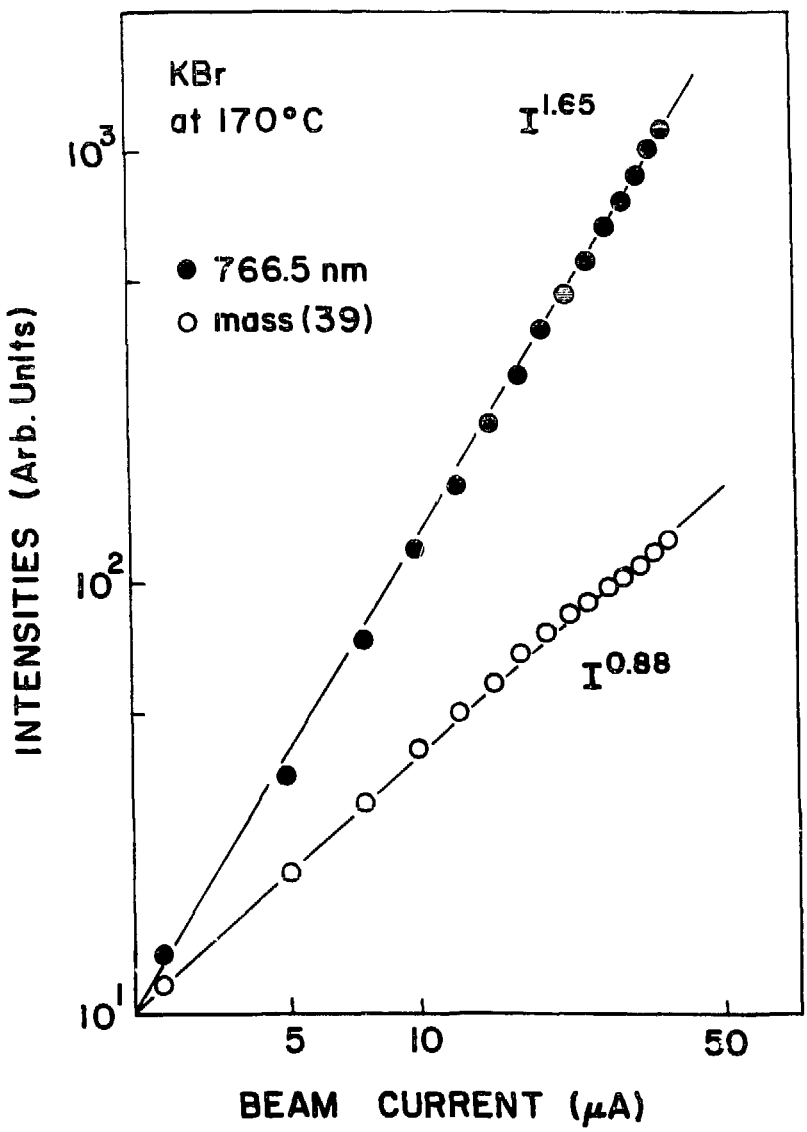

Fig. 4. Beam current dependence of the ground-state and the excited-state potassium atom intensities from KBr at 443°K.

the case of electron bombardment most of the observed radiation originates in gas-phase collisions above the surface. On the other hand, Tolk et al.[13-15] observed a linear dependence of the optical emission intensities on electron beam current from NaCl at T = 733°K and have suggested that the excited states are produced by electronic excitation of surface alkali atoms.

Postawa et al.[12] suggest that under certain circumstances (low sample temperature, high electron current) the thermal evaporation of the alkali metal overlayer totally controls the sputtering, and the atom yield is independent of electron current. This would lead to a linear dependence of photon intensity vs current for the gas-phase excitation. Our optical emission data in region I for KBr at room temperature appear to be consistent with the above, although the Auger data in region I seem to show good stoichiometry of our sample surface. Tolks' linear dependence at T = 723°K is also difficult to understand. Clearly, stoichiometry of the alkali halide surface seems to be involved as well as radiation damage in regions II and III of our room-temperature data. However, more work needs to be done in order to interpret our room-temperature data in region I.

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