

Comment on

Regenerative soot as a source of broad band VUV light by Shoab Ahmad

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Abstract. The analysis and interpretation of the data presented by S. Ahmad [Eur. Phys. J. D **22**, 189 (2003)] are shown to be inconsistent, and are in conflict with established atomic physics.

PACS. 32.70.Fw Absolute and relative intensities – 34.50.Dy Interactions of atoms and molecules with surfaces; photon and electron emission; neutralization of ions – 34.80.Dp Atomic excitation and ionization by electron impact

The paper by S. Ahmad [Eur. Phys. J. D **22**, 189 (2003)] raises a number of very interesting points. They relate to the general questions of what peculiarities of specific light sources might yield new scientific insight into atomic or plasma physics processes, and whether they might have promising technical applications. If a number of the claims made in the paper could be corroborated, the sooty discharge used would make a very interesting device, indeed. However, as I shall line out below, there is some doubt concerning the corroboration of the claims from the material and discussion presented by S. Ahmad.

The author operates a hollow cathode discharge. Gas mixing effects in such discharges have produced astonishing effects on certain line intensities. It would be interesting to find out whether the sooty discharge that the author used also shows such interesting peculiarities. However, the discharge is not characterized in sufficient detail as might permit a scientific reproduction of the work. Instead, a number of arguments are brought forward to promote the case. Unfortunately, the reasoning of various of these arguments as far as they relate to spectroscopy and the observation of decays from long-lived atomic levels contradicts accepted views and established knowledge. This on its own might not be such a bad thing, if the unorthodox treatment was scientifically consistent and sound.

The author highlights the role of metastable levels, claims the appearance of multiply charged ions in the hol-

low cathode discharge as well as the observation of high intensity intercombination lines of both C II and C III, and calls his discharge a broad band VUV light source in the title and the abstract. His Figure 1, however, shows light emission mostly in the visible and UV, and hardly anything below 200 nm. I assume that the light path was not evacuated. Therefore the data barely reach the range defined as vacuum ultraviolet (VUV), and encompass everything that the air in the light path has let pass and the unspecified detector could see. Moreover, the role of “regenerative soot” in the working of this light source is never explained. In my discussion of the aforementioned central points, I shall use only estimates of various entities, instead of presenting formulae and seemingly exact factors. This simplification may be justified by the fact that Ahmad’s interpretation and mine differ by about six orders of magnitude.

Ahmad gives among the properties of the discharge an electron density of $n_e \approx 10^{10-12} \text{ cm}^{-3}$, which seems compatible with the densities of other light sources (quoted earlier in the paper) in which intercombination transitions have been seen, that is, for example, from planetary nebulae and tokamak discharges. However, the gas pressure in the sooty discharge was 0.1 mbar (text pointing to Fig. 1 of Ahmad’s paper) or 0.6 mbar (caption of Fig. 1). This puts the particle density in the discharge to about 10^{15} cm^{-3} , and that is much more than in any of the other light sources. Assuming geometric cross-sections, the collision time of the molecules in ambient air is of the order of one nanosecond. In the gas discharge discussed here,

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the pressure is 3 to 4 orders of magnitude lower, and the collision time thus increases to microseconds or tens of microseconds. This is some 5 to 10 orders of magnitude less than the collision times, for example, in planetary nebulae where long-lived excited levels have a chance to radiatively decay instead of being collisionally quenched [1–4], and where thus decays of even very-long lived levels have been observed [5].

In the laboratory, pressures as low as 10^{-9} mbar have a notable effect on the apparent lifetime of levels with 20 ms lifetime [6, 7]. Ahmad used a pressure that was 8 orders of magnitude higher. Under such poor vacuum conditions, the radiative decay fraction of a long-lived level is minute, and collisional quenching dominates. When, more than a hundred years ago, vacuum production was a problem, spectroscopic light sources were flames and arcs, and the spectra showed only neutral and singly charged atoms. Only with improving vacuum technology the higher charge states appeared, because charged particles now fared a sizeable chance of gaining sufficient energy (from external fields) between collisions so that collisions might be energetic enough to ionize atoms, and ionizing collisions be frequent enough in their competition with the much more frequent neutralizing collisions. It would be very useful if a hollow cathode discharge could be run in a way that produces as many doubly or even triply charged ions as Ahmad claims. The experience with ion sources for accelerators speaks differently.

However, what proof is there for the presence of these ions in Ahmad's discharge? All the evidence supposedly is derived from spectra like the ones shown in the paper, recorded with Ne as a carrier gas. The spectroscopic equipment is not mentioned, nor is any quantification of the spectral calibration or resolution given. All labeled lines in the short wavelength part of the displayed spectrum are identified with carbon in its various ionization stages, and lines in the long wavelength part are indicated as belonging to Ne I. The line identifications appear to be based solely on wavelength coincidences with entries in spectral tables. Considering the line widths in the spectra shown, the not specified reliability of the experimental wavelength scale, the number of lines available in spectral tables, and the incomplete analysis (about half of the lines are not identified, which, by the way, is quite common), the question arises of how certain the line identification process is. This is not a problem that pertains to this work only; spectroscopists face this challenge over and over again. Additional clues are needed, like multiplet patterns of line splittings and relative intensities, systematic checks of the consistency of excitation functions, or the completeness of decay patterns. For example, if a decay to a level of a given principal quantum number n is observed, usually also the decays to lower levels appear, as well as subsequent decays in the same decay chain. Also, if a level with n is reached, one checks for excitations of lower-lying levels, because these normally (for collisional or thermal excitation) would be populated more prominently. Exceptions from this rule would indicate selective excitation processes. Working through Ahmad's line la-

bels, I find rather spurious assignments, and no systematic consistency. In the light of the standard procedure, the line identifications given (including the lines that are simply labeled by a spectrum number in Ahmad's Fig. 1) appear to be fortuitous. Using these to construct the decay scheme in Ahmad's Figure 3 seems daring. If the line identifications were correct, the atoms would show highly selective excitation processes only, including selective deexcitation that is in conflict with quantum mechanics. In several cases the identified line leads to displaced terms (usually rather weak branches), and the major decay branch is missing.

On these line identifications hinges a central point of Ahmad's paper, the appearance of bright lines that are claimed to represent intercombination transitions in C II and C III. The decay rates of the long lived levels in both ions have been measured [8–10, 12]. These measurements all used ultra-high vacuum conditions and ion traps in order to be able to detect the low-rate radiative decays. In fact, in the storage ring experiments, the vacuum was better than that in Ahmad's discharge by about 10 orders of magnitude, and even at that level, variations of the vacuum pressure were shown to influence the raw data. In C II, three fine structure levels, of lifetimes 8 ms, 104 ms, and 22 ms, contribute to the intercombination transition multiplet at 233 nm. These lifetime results from a heavy-ion storage ring [10] have been corroborated by recent calculations [11]. In C III (intercombination line near 191 nm), recent experiment [12] and theory [13] are both very precise, and disagree only at a very high level of accuracy. It is not very likely (by some 6 orders of magnitude) that these transitions would be seen at all under the vacuum conditions of Ahmad's discharge.

A third intercombination line is brought up by Ahmad to explain a line at 219.6 nm. This would be the C II $2s2p^2\ ^2S_{1/2}-2p^3\ ^4S_{3/2}$ transition, with a transition probability of (quoted) $5.1\ \text{s}^{-1}$. However, the upper level of this transition is not long lived. The dominant decay branch is a regular, fully allowed, electric dipole transition, which outweighs the weak intercombination branch by about 6 orders of magnitude. If this intercombination transition was the real origin of the line in Ahmad's spectrum, there would be the allowed line to search for, at a million times higher brightness.

Ahmad recognizes that "the two intercombination transitions are 6–7 orders of magnitude more intense compared with the allowed transitions". The obvious conclusion should have been that the lines seen are not the intercombination transitions in the claimed atomic ions at all. This conclusion, evidently, would invalidate the very substance of the paper.

It is not very likely that the atomic collision processes in Ahmad's hollow cathode discharge work so differently from what spectroscopists have been assuming for about a century. Is there anything else that would explain the lines seen in the spectra shown, apart from the unknown wavelength calibration, non-existent efficiency calibration, and unspecified gas purity? If the contributions from atoms and atomic ions (of carbon or neon) can be clarified after putting the spectroscopic basis straight, and if the possible

role of magnetic fields (which are not common to conventional hollow cathode lamps, but are present here) has been sorted out [14], then what is the role of the soot in this “sooty” discharge? Could clusters of carbon be involved, might they have allowed transitions where atoms only have spin forbidden transitions? The data give rise to many open questions that possibly are worth pursuing.

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