Electron excitation energy transfer in mixed krypton-xenon clusters

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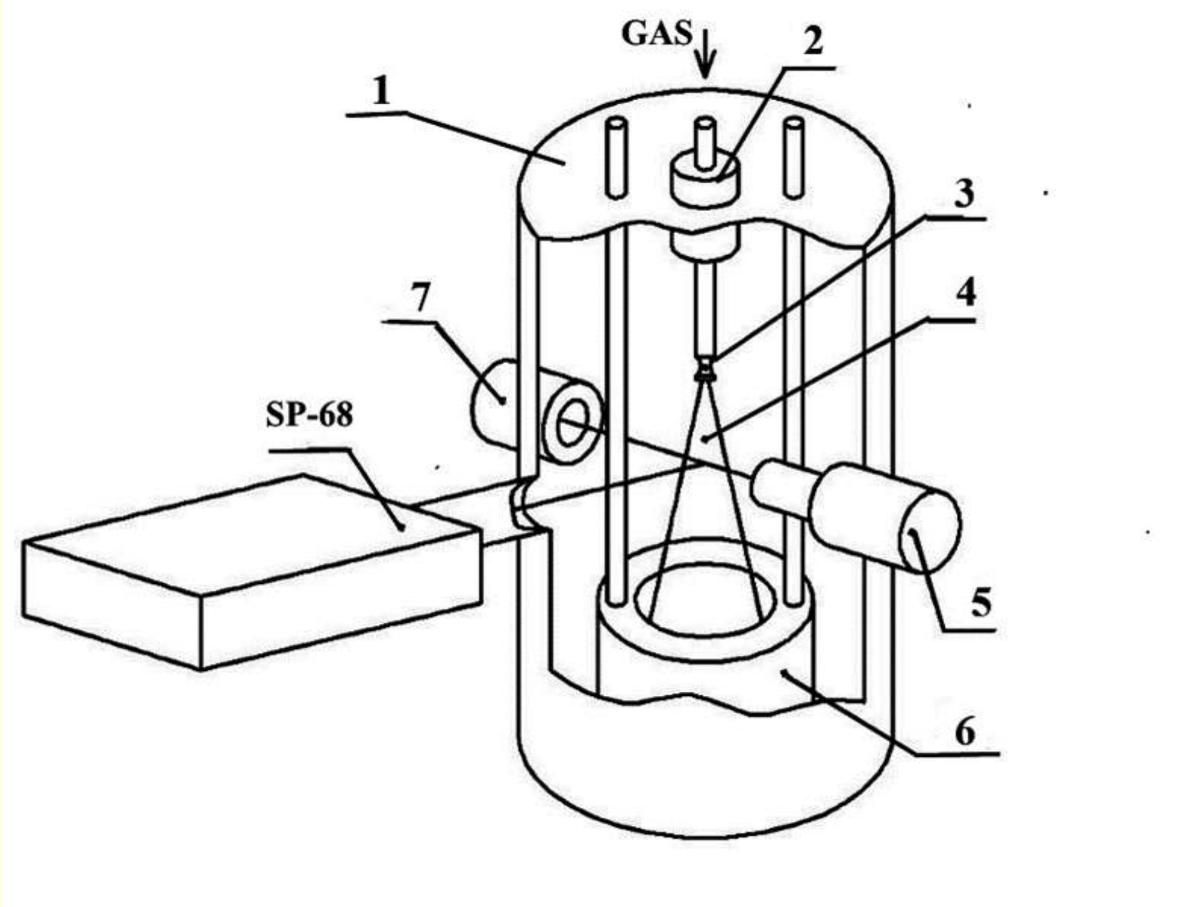
INTRODUCTION

The spectroscopic study of the emission spectra of heterogeneous rare-gas clusters is of interest for several reasons. Firstly, they may provide information regarding the formation of new heteronuclear diatomic excited rare gas molecules in clusters. Secondly, studying the formation of excited impurity two-atomic molecules and their subsequent radiative decay provides information on the medium-induced intramolecular vibrational relaxation in heterogeneous clusters. It is also interesting to understand the mechanisms of electron energy transfer from the excited two-atomic host molecule to impurity states. Previous studies of heterogeneous Ar-Kr clusters [1] have demonstrated that the cluster size, structure, number of impurity atoms, and location significantly influence the energy transfer processes of electronic excitations in two-component clusters.

METHODS

The two-component Kr-Xe clusters are formed in a supersonic gas jet exiting the vacuum. The size of the clusters can be varied by adjusting the gas pressure (P_0) and the temperature (T_0) at the nozzle input. The clusters are excited by electrons with an energy of 1 keV and a 20 mA current. The cathodoluminescence (CL) spectra were recorded in the wavelength interval between 130 and 200 nm.

Experimental setup



1 - generator housing; 2 - heat exchanger; 3 -supersonic conic nozzle; 4 - supersonic gas jet; 5 - electron gun; 6 - cryogenic pump; 7 - Faraday cup; SP-68 - spectrometer-monochromator

The average size (in at/cl) for two-component Kr-Xe clusters and the enrichment coefficient of the Kr-Xe system were determined by the electron diffraction method [2].

[1] Yu. S. Doronin; V. N. Samovarov; E. A. Bondarenko, Low Temp. Phys. 32, 251 (2006) https://doi.org/10.1063/1.2178482

[2] O. P. Konotop, S. I. Kovalenko, O. G. Danylchenko and V. N. Samovarov, J. Clust. Sci. 26, 863 (2015) DOI:10.1007/s10876-014-0773-6

*[3] E.T. Verkhovtseva, E.A. Bondarenko and Yu. S. Doronin, Low Temp. Phys. **30**, 34 (2004) [Fiz. Nizk. Temp. **30**, 47 (2004)] https://doi.org/10.1063/1.1645153*

RESULTS

The average Kr-Xe cluster size $\bar{N}_{\mathrm{calc}}$ (in at/cl) is determined as:

$$\bar{\boldsymbol{N}}_{\mathbf{calc}} = 19{,}5\left[\boldsymbol{k}_{\mathbf{Kr}}^{1,8} + \frac{\boldsymbol{C}_{\mathbf{Xe}}^{\mathbf{gas}}}{\boldsymbol{C}_{\mathbf{CR}}}\left(\boldsymbol{k}_{\mathbf{Xe}}^{1,8} - \boldsymbol{k}_{\mathbf{Kr}}^{1,8}\right)\right]\left\{\left(\frac{0{,}74\boldsymbol{d}}{\tan\boldsymbol{\alpha}}\right)^{0,85}\frac{\boldsymbol{P}_0}{10^3\boldsymbol{T}_0^{2,29}}\right\}^{1,8}$$

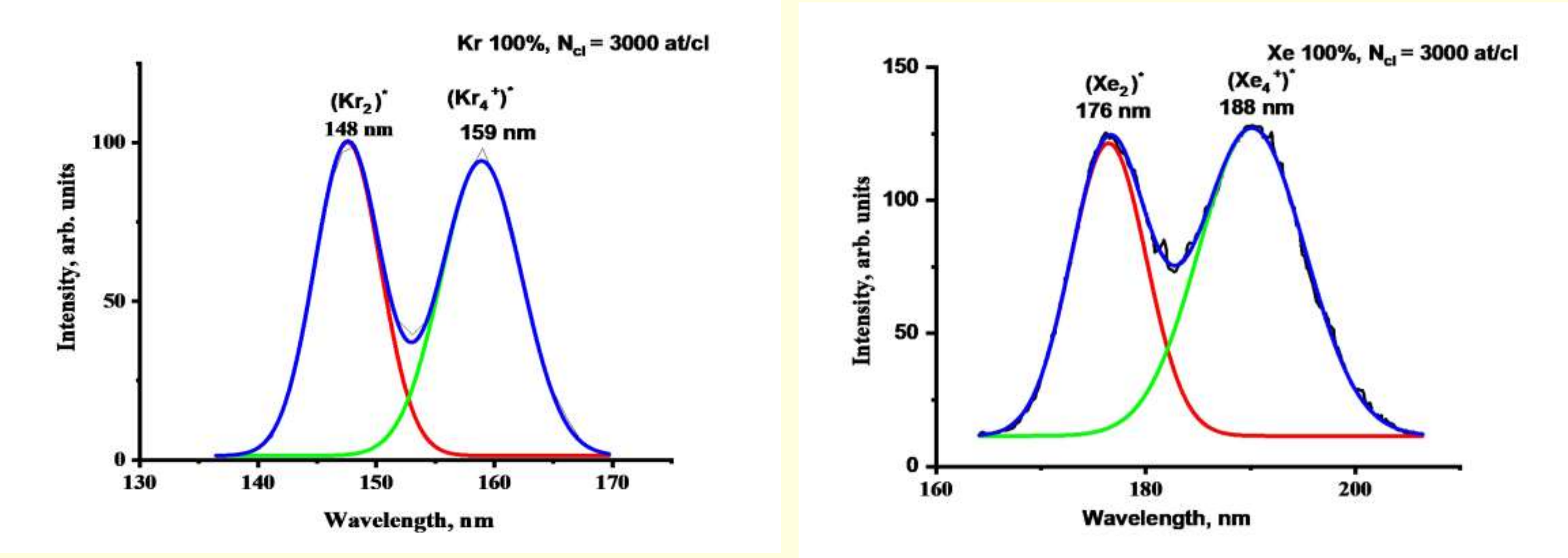
where $\mathbf{P_0}$ is the total gas mixture pressure, $\boldsymbol{k}$ is the characteristic constant of the gas, $\boldsymbol{C_{Xe}^{gas}}$ is the volume fraction of xenon in the gas mixture, $\boldsymbol{C_{CR}}$ is the volume fraction of xenon in the mixture at which and above which only homogeneous xenon clusters condense, regardless of the values of pressure P_0 and temperature T_0, $\boldsymbol{d}$ –critical diameter of the conical nozzle, $\boldsymbol{2\alpha}$ **-** the total cone angle.

The enrichment factor for two concentrations of Xe in the gas mixture, 1 and 2% is:

$$\boldsymbol{C}_{\mathbf{Xe}}^{\mathbf{cl}} = \boldsymbol{exp}\left[\frac{\boldsymbol{\beta}}{\boldsymbol{4\pi}}\left(\boldsymbol{C}_{\mathbf{CR}} - \boldsymbol{C}_{\mathbf{Xe}}^{\mathbf{gas}}\right)\times\left(\frac{\mathbf{1}}{\boldsymbol{R}} - \frac{\mathbf{1}}{\boldsymbol{R}_{\mathbf{CR}}}\right)\right]$$

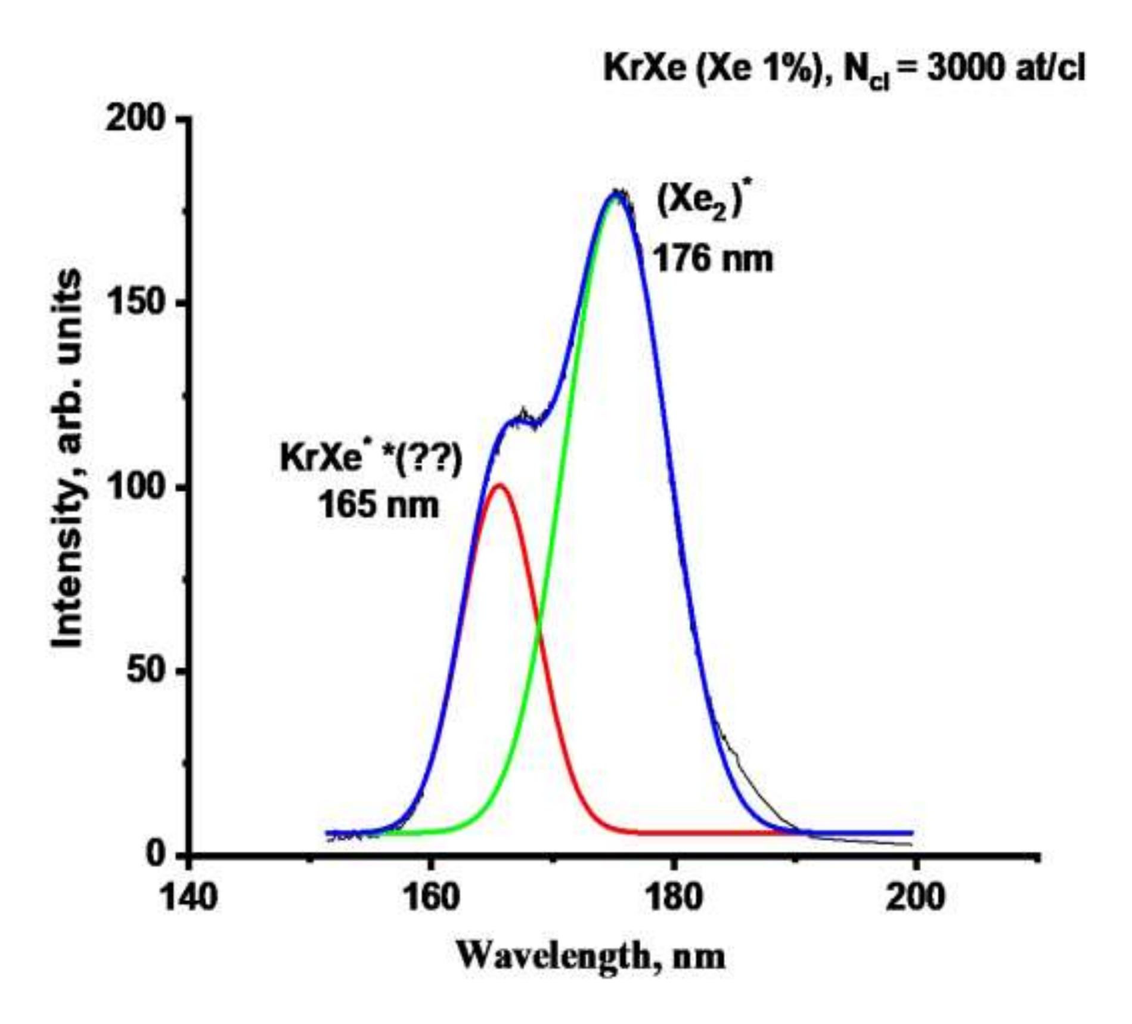
where $\boldsymbol{\beta}$ is the constant of proportionality, $\boldsymbol{R}$ –radius of the cluster, $\boldsymbol{R_{CR}}$ **-** critical radius of the cluster, above which krypton condenses and mixed Kr-Xe clusters are formed.

One-component Kr and Xe clusters



CL spectra of the one-component Kr and Xe clusters (N_{cl} ≈ 3000at/cl). The spectra contain well-known emission bands of neutral excimers Kr_2^* and Xe_2^* and charged complexes $(Kr_4^+)^*$ and $(Xe_4^+)^*$ [3].

Two-component Kr-Xe clusters



The CL spectrum of Kr-Xe clusters measured at xenon concentration of 1% in the gas mixture. The registered 165 nm band is emitted from the cluster by the Kr-Xe exciplex. N_{cl} ≈ 3000at/cl. The concentration of xenon in the Kr-Xe clusters is 3.6%. The emission bands of neutral Kr_2^* excimers and charged complexes $(Kr_4^+)^*$ are not observed in the spectrum.

CONCLUSIONS

The obtained result indicates that at a concentration of xenon in the gas mixture at the nozzle inlet of 1% at P_0 = 0.125 MPa and T = 215 K, only two-component Kr-Xe clusters with a notable enrichment of xenon atoms are formed in the jet. At the same time, an Xe core with a diffuse boundary with the krypton shell is formed in the clusters. The complete quenching of the Kr_2^* and $(Kr_4^+)^*$ molecular continua characteristic of krypton clusters in the Förster-Dexter process leads to the appearance in the spectrum of an intense molecular continuum emitted by the Xe_2^* molecule. The absence of $(Xe_4^+)^*$ emission in the spectrum can be attributed to the low probability of ion complex formation in a xenon core of limited size. The continuum at λ=165 nm, registered for the first time in the emission of Kr-Xe clusters, has a significant shift compared to the solid and liquid states. We propose that this continuum is emitted by the Kr-Xe* exciplex from the disordered icosahedral cluster shell.

Further studies on these topics are in progress…