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COMMENT ON "EXCITON LIFETIMES IN ELECTRON BEAM EXCITED CONDENSED PHASES OF ARGON AND XENON"

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Comment on "Exciton lifetimes in electron beam excited condensed phases of argon and xenon"

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In a recent article, Keto, Gleason and Soley[1] reported decay times for the 1750 Å emission band of solid xenon in the temperature range of 65 K to 136 K. The crystals had been excited by electrons of 200 keV at a current of 10^{-2} to 10^{-6} A. In Fig. 1 we gathered decay times for solid Xe in the temperature range of 4 K to 200 K published by Hahn et al.[2], Kink et al.[3] and Keto et al.[1], and which includes unpublished results of Hahn et al.[4]. Fig. 1 shows that the decay time of the 1750 Å band of solid Xe is strongly temperature dependent. It increases by about three orders of magnitude in going from 100 K to 5 K. The decay times of solid Kr show a similar behavior[5]. The statement of Keto et al.[1] that the decay time of solid Xe is temperature independent is incorrect for temperatures below 50 K.

Keto et al.[1] mention the results of Hahn et al.[2]. They do not discuss the obvious temperature dependence of the decay time but point out surface sensitivity after excitation with 300 eV electrons used by Hahn et al.[2]. Hahn et al. have been aware of this problem. They observed a decrease of decay time with increasing contamination and explained it as a result of surface quenching. The additional channel introduced by surface quenching will only reduce the decay times but contrary to Keto et al.[1] cannot account for the longer lifetimes at low temperatures which have been found by Hahn et al. Therefore, the increase of the decay times at

low temperatures to only 1 μsec after electron excitation[2] instead of more than 10 μsec after optical excitation[3,4] is attributed to surface quenching.

As an explanation of the decay time of the 1750 Å band of Xe, Keto et al.[1] propose mixing of the $^3\Sigma_u$ and $^1\Sigma_u$ state. The decay times calculated from collisions with phonons at a collision frequency derived from the gas phase are too long. Next, they discuss collision with free electrons at an electron density given by their beam currents. Finally, they estimate recombination times for ions and electrons. To test their models they propose decay time measurements after light excitation.

Decay times obtained by light excitation have been published[3] and are supported by our unpublished results[4]. In both experiments the samples had been excited just above the energy of the n = 1 exciton (8.4 eV) which assures a large penetration depth of the light and consequently bulk excitation. The used excitation energies (8.8 eV, Kink et al.[3], 8.8 eV Hahn et al.[4]) are far below the ionisation energy (9.33 eV), thus neither electron collisions nor recombination of electrons and ions can influence the decay times. The results (Fig. 1) agree with the electron excitation experiments carried out at temperatures above 20 K.

This shows clearly that the decay times cannot be explained as a result of collisions with electrons and of ion electron recombination times. The key to an understanding is to consider the strong temperature dependence. This temperature dependence therefore supports the attempts of Hahn et al.[2] and Kink et al.[3] to interpret the decay times by a phonon mixing of molecular states in the symmetry of the solid.

References

1) J.W. Keto, R.E. Gleason, F.K. Soley
J.Chem.Phys. 71, 2676 (1979)

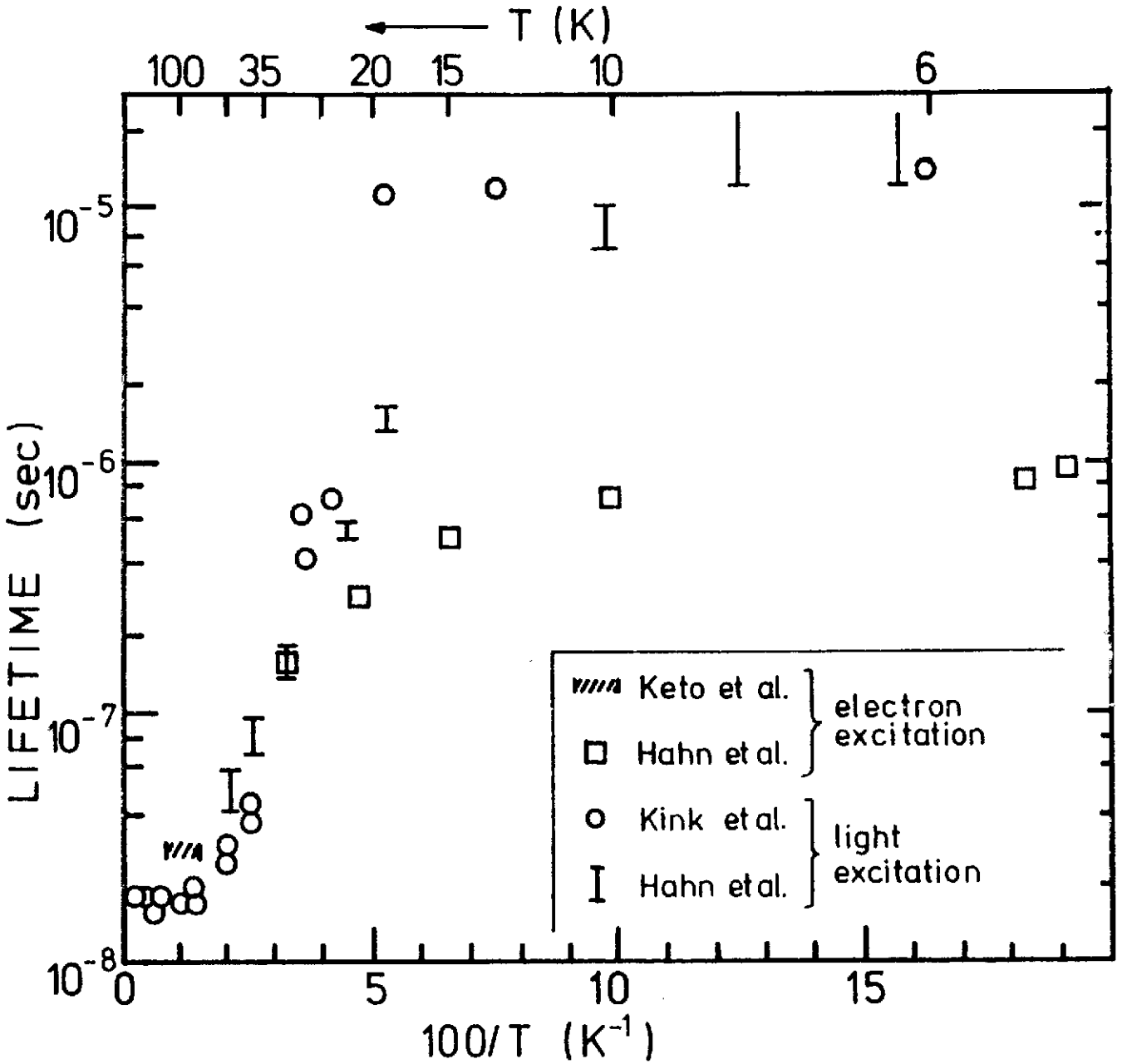
2) U. Hahn, N. Schwentner, G. Zimmerer
Opt.Commun. 21, 237 (1977)

3) R. Kink, A. Lõhmus, M. Selg and T. Soovik
phys.stat.sol.(b) 84, K61 (1977)

4) U. Hahn, N. Schwentner, G. Zimmerer, unpublished.
Condensed films of solid Xe have been excited by monochromatized synchrotron radiation at 8.8. eV. The emitted light has been recorded at 1770 Å by means of a second monochromator. For the setup and the decay time measurements see: Nuclear Instrum. and Methods 152, 261 (1978)

5) G. Zimmerer, J.Luminescence 18/19, 875 (1979)

Fig.1 The temperature dependence of the decay time for the 7.2 Xenon emission band.


T (K)
100
35
20
15
10
6
10^{-5}
10^{-6}
10^{-7}
10^{-8}
LIFETIME (sec)
0
5
10
15
100/T (K^{-1})
Keto et al.
Hahn et al.
electron excitation
Kink et al.
Hahn et al.
light excitation