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LASER SPECTROSCOPY OF QUANTUM WELL AND SUPERLATTICE
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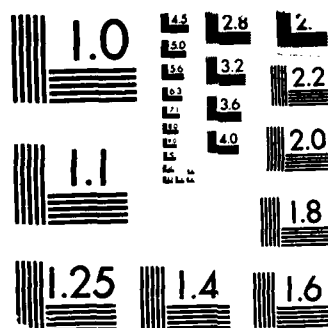
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LASER SPECTROSCOPY OF QUANTUM WELL
AND SUPERLATTICE STRUCTURES

by

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A. Research Programme

The research described in this report aims to understand the effects of disorder on the lineshapes of exciton luminescence in quantum well (QW) materials. The low temperature photoluminescence (PL) and photoluminescence excitation (PLE) spectra of the excitonic transition in QW material are a sensitive test of interfacial quality.^[1] Contributions to the linewidth arise from homogeneous (lifetime) and inhomogeneous (disorder) effects.^[2] In a typical 50 Å well in GaAs, a linewidth at low temperatures of a few meV compares to a typical linewidth about one hundred times smaller in bulk material. Weakly bound impurities, such as the residual C_{AS} acceptor, may lead to unresolved structure in the emission line, thus broadening it.^[3] In addition, since most samples are multiple quantum wells, say 50 individual wells grown sequentially, there is a macroscopic contribution to the inhomogeneous linewidth if the individual wells differ from each other in mean dimension.^[4]

In QWs grown to date, inhomogeneous effects dominate the observed width. The QW exciton is an optical probe approximately 300 Å across which samples the environment in which it is formed or in which it decays. The sampling yields, through the photon energy for exciton formation/destruction and the homogeneous width, detailed information about the disorder in the QW. Since different parts of the exciton line are associated with different environments, a fine line tunable laser can be used to probe the disorder by means of resonant excitation of sub-well environments.



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B. Sources of GaAs/AlGaAs materials

Two samples of nominal well width 55 Å (20 monolayers) were obtained from P. Dawson of Philips Research Laboratories (PRL) at Redhill. The sample KLB155 (KLB219) is an undoped multiple quantum well formed by alternating 60 (55Å) layers of undoped GaAs with much thicker (175 Å) barrier layers of $\text{Al}_{0.35}\text{Ga}_{0.65}\text{As}$. Both samples show prominent narrow low temperature PL bands near 1.60 eV but are distinguished by the value of the (misnamed) 'Stokes' shift'[5] between the peak of the luminescence band and the corresponding excitation peak. At very low temperatures, recombination involves mainly the $n=1$ electron quantum well state with the split-off $n=1$ heavy hole quantum well state. We shall refer to this as the E_{1h} recombination, following R.C. Miller's notation.[6] For sample KLB155, $E_{1h}(\text{PLE}) - E_{1h}(\text{PL}) \sim 2$ meV while for sample KLB219, the 'Stokes' shift' is $< \frac{1}{2}$ meV.

C. Experimental details

Samples were cooled either by a liquid-helium exchange gas cryostat (Oxford Instruments MD4) or in a closed-cycle helium refrigerator (Cryophysics) equipped with optical tails. Luminescence was excited using an Ar^+ pumped dye laser (Spectra Physics) or a temperature-tuned AlGaAs laser diode (Liconix) and detected using a GaAs phototube (Hamamatsu) at the exit slit of a 1 m grating monochromator. Excitation spectra were obtained 'point-by-point' using the diode laser or quasicontinuously using a Pyridene dye laser. Luminescence peaks are monitored exclusively in these experiments. Excitation intensities were typically 10 mW/cm^2 .

D. Experimental details

(i) Luminescence lineshape temperature dependence function of temperature.

The integrated intensity, the peak position and the halfwidth of E_{1h} luminescence from sample KLB155 show the temperature dependences expected from previous work. The intensity is roughly constant from 5 K to 25 K and then decreases rapidly in the temperature range 25 K - 100 K. At 170 K the total intensity is about 10% of the low temperature value. The peak position follows the GaAs bandgap decreasing by ~ 2 meV/K in the temperature range from 75 K to 170 K. The linewidth increases approximately linearly with a temperature above ~ 15 K with a slope of ~ 0.5 meV/K (cf Boltzmann's constant = $.87$ meV/K). The low-temperature PL half-width is ~ 6 meV. Closer examination of the line for this sample shows that it consists of two major components. Excitation at 1.635 eV favours the weaker high-energy component as confirmed by the excitation spectrum: the majority light hole excitation E_1 peaks at 1.624 eV and the minor light hole peaks at 1.634 eV. The heavy hole luminescence bands corresponding to these excitations peak at 1.601 eV and ~ 1.607 eV. The difference between the major and minor peaks is emphasised by the temperature dependence of the semi-half widths of the luminescence band. The low-energy side of the line is observed to broaden much more slowly than does the high-energy side. [See Figure 1(a).] In contrast, the luminescence from sample KL219 is unstructured. The E_{1h} excitation and luminescence lines have peaks within 0.5 meV of each other at 1.583 eV. The excitation line excitation and luminescence lines have peaks within 0.5 meV of each other at 1.583 eV. The excitation line E_1 occurs at

1.603 eV. The luminescence band broadens only very slowly with increasing temperature and has a value of 5.1 meV at $T \leq 85$ K. [Figure 1(b)]

(ii) *Resonant excitation spectra: strain-induced 'Stokes' shift*

Luminescence and quasicontinuous excitation spectra in the region of the E_{1h} line for both samples are shown in Figure 2. Note the different energy scales indicated by the 2 meV energy bar on the figures. Luminescence and excitation peaks for each sample show the so-called 'Stokes' shift' but that for sample KLB155 is the larger (2meV). This is made clearer by specifying the efficiency of excitation (relative to the excitation peak) at the peak of the luminescence band. For sample KLB155, this 'figure of merit' is only 70% while for KLB219 it is 94%. (For a sample with zero 'Stokes' shift', the figure would be 100%.) The excitation spectra shown in Figures 3a,b,c were obtained point-by-point using a Liconix Diolite AlGaAs diode laser as excitation source. This source emits temperature-tuned radiation of very narrow bandwidth (measured to be ≤ 50 MHz using a scanning Fabry Perot interferometer). For these runs sample KLB219 was mounted in such a way as to induce strain in the sample upon cooling to cryogenic temperatures. Severe strain was induced by coating the rear face of the sample with aquadag and less severe strain (Figures 3a,b) by using a thin coat of G.E. varnish. For comparison, a free-standing sample mounted in the same cryostat showed only a very small 'Stokes' shift'. Figures of merit for the strained and very strained sample are 40% and 20% respectively. The very strained sample exhibits a strain-induced 'Stokes' shift' of ≥ 4 meV.

E Discussion of results

Despite initial appearances, samples KLB155 and KLB219 are very different. KLB219 shows a very small 'Stokes' shift' (in the absence of strain), a moderate low temperature PL linewidth, no discernible structure in the E_{1h} emission line and a very weak dependence of the linewidth on increasing temperature. Sample KLB155 shows a moderate 'Stokes' shift' of 2 meV. Values for 55 Å QWs of up to 6 meV have been reported in the literature.[7] The band shows discernible structure with a partially resolved higher energy component having $E_{1h}(\text{lum}) = 1.607$ eV and a resolvable light hole excitation peak at 1.634 eV (cf. 1.601 eV and 1.624 eV). The broadening of the luminescence band with temperature is very marked for this sample. The minor peak appears to thermalise with the major peak and also to broaden rapidly, suggesting that minority regions of the well having a smaller mean well-width may be responsible for this behaviour. The increased $E_{1h}-E_{1\ell}$ separation and E_{1h} energy appear also to be consistent with this interpretation, although it is impossible to obtain a numerical agreement with theory in this case. The difference in temperature-broadening of the luminescence requires an explanation. Broadening with kT occurs at a rate of 3.2 meV per 100K, as observed for the total linewidth in sample KLB155. An increase smaller than this value might imply that the sample is not 'feeling the heat', i.e. that the phonons with which the exciton interacts are in fact fairly hard to excite. What are we to make of the different behaviour of sample KLB219 for which the exciton linewidth hardly appears to vary in the temperature range from 4 K to 100 K? We assume that the linewidth is $\Gamma = \Gamma_0 + \Gamma_w(T)$ where $\Gamma_w(T)$ reflects the phonon occupation through the relation

$$\Gamma_w(T) = \Gamma_w / [\exp(\hbar\omega/kT) - 1]$$

If we further assume that $\hbar\omega/k \sim 420$ K, as expected for a (3-D) GaAs LO phonon, the thermal broadening is expected to be $0.014 \Gamma_w$ at 100 K and $0.31 \Gamma_w$ at 300 K. We then consider that if the exciton linewidths are broadened by the same LO phonons in each case, then the broadening factor Γ_w is larger for the minority exciton in sample KLB155 by at least an order of magnitude. It is difficult to imagine why this might be so. If the differences reflect gross differences in interfacial quality, why is this not reflected in the low-temperature linewidth which is due entirely to such inhomogeneous effects?

The shift in the peak of the excitation with increasing strain on sample KLB219 is not too surprising. Axial strain leads to a deformation splitting of the valence band of ~ 1 eV per unit strain in bulk GaAs and similar values are expected in strained QWs. Weisbuch et al.[8] have reported exciton absorption shifts due to strain upon chemically removing the GaAs substrate on a similar sample. It is not clear from Miller[9] whether the exciton luminescence peak has followed the absorption peak when strain is present. Since we are measuring the luminescence excitation point-by-point in our experiments, the strain-induced shift between luminescence and excitation peaks cannot be mistaken. It is also perhaps worth noting that there is no evidence of enhanced Rayleigh scattering[10] when the excitation light is resonant with the exciton in our CW measurements.

In the strained sample, excitons appear to form more readily at a higher energy than that at which they most readily decay. It is as if the 'Stokes' shift' represented a kind of strain-induced binding energy. However, it

would be incautious to say too much about preliminary work. The experiments must be repeated with a calibrated stress applied to the sample. More quantitative work must be done to disentangle, for example, the contribution to this effect of homogeneous and inhomogeneous strain field. In our experiment a fairly long but quite narrow strip ($\sim 2 \text{ mm} \times 10 \text{ }\mu\text{m}$) of sample contributes to the observed signal. Suitable masking of the fluorescent image will allow a strain image of the sample to be built up in future experiments.

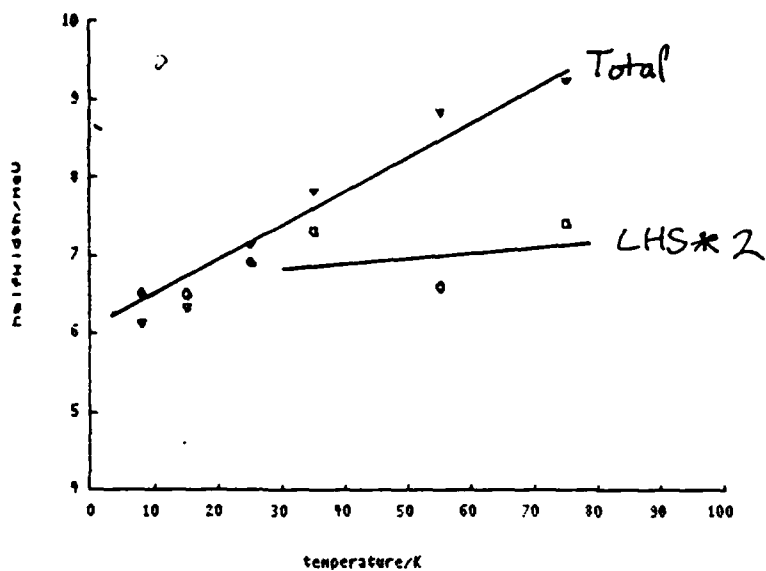
B Prospects for future work

The work described here has served the purpose of familiarising the writer with the optical properties of GaAs quantum wells. Several strands may be teased out in further work. Firstly, the effects of magnetic fields on the circular polarisation properties of the wells must be tackled with a reliable cryostat/superconductive magnet. The projected measurements of spin-dependent emission and ODMR depend upon a good grounding in MCP. Secondly, the effect of strain on QW luminescence and excitation is worth pursuing in a quantitative fashion. This study, interesting in itself, will also throw light on those materials with inbuilt strain, viz. the strained-layer systems like Zns , $\text{ZnS}_x\text{S}_{1-x}$ and $\text{Si/Si}_x\text{Ge}_{1-x}$. A number of recent papers[11] have described exciton transitions in QW grown with interruptions of reactants between layers. Very narrow lines are obtained in these transitions, with values as low as 1 meV for $\sim 57 \text{ }\text{\AA}$ (20 monolayer) wells.[12] In such transitions, satellite structures have been attributed to recombination in parts of the well differing in thickness by a single monolayer from the mean width yet having spatial extents large compared to the

exciton radius. It would be very interesting indeed to repeat the present measurements described here on material which allowed the exciton environments to be distinguished so clearly.

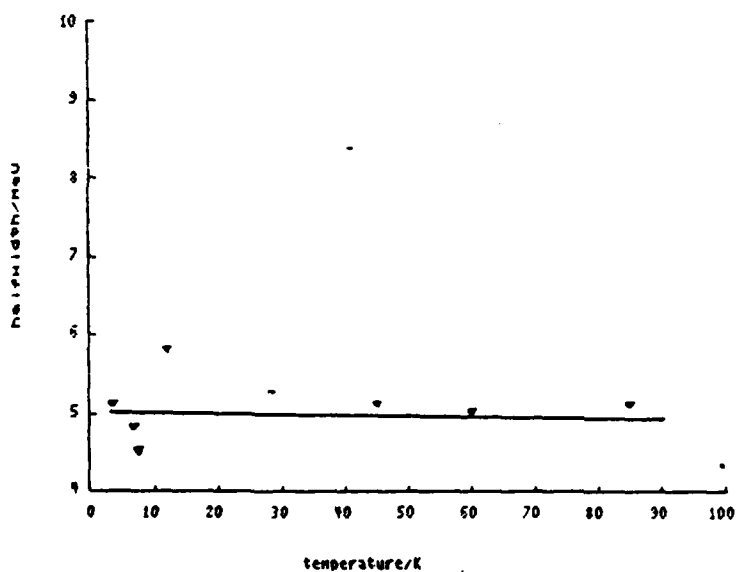
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Halfwidth as function of temperature for sample lib155

Figure 1 (a)



Halfwidth as function of temperature for sample lib215

Figure 1 (b)

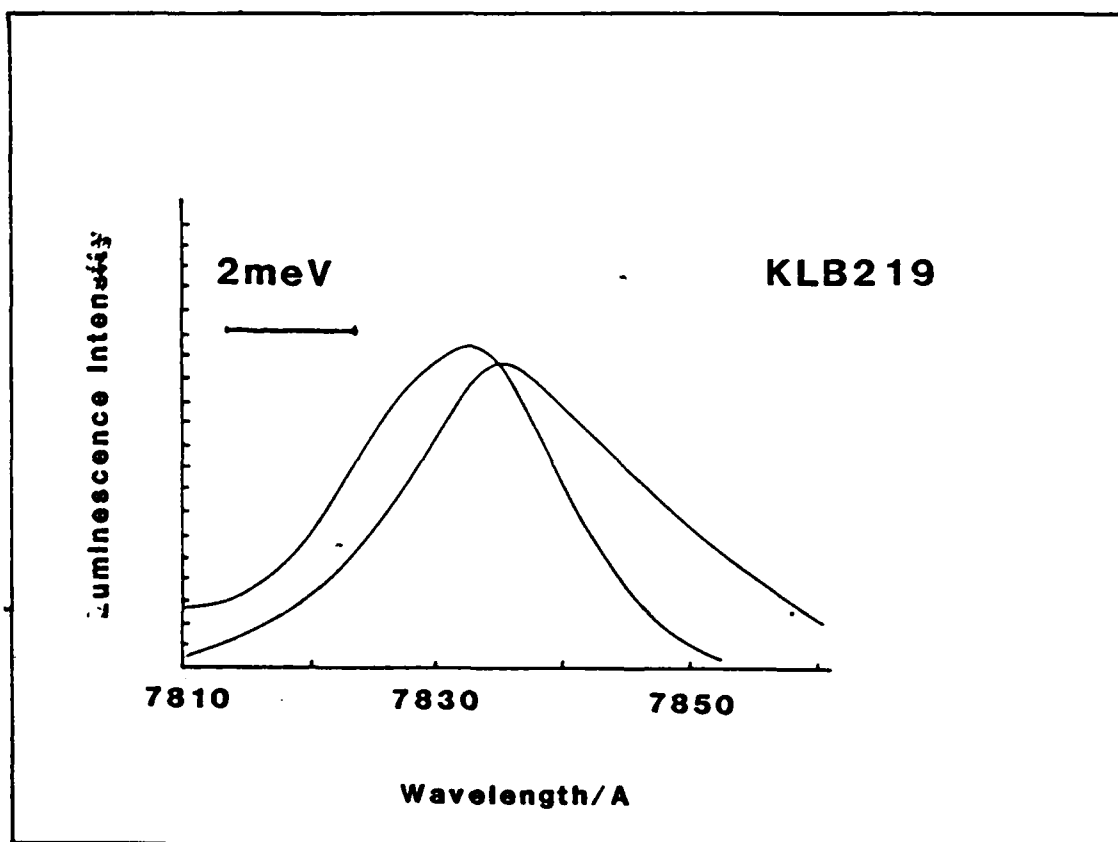
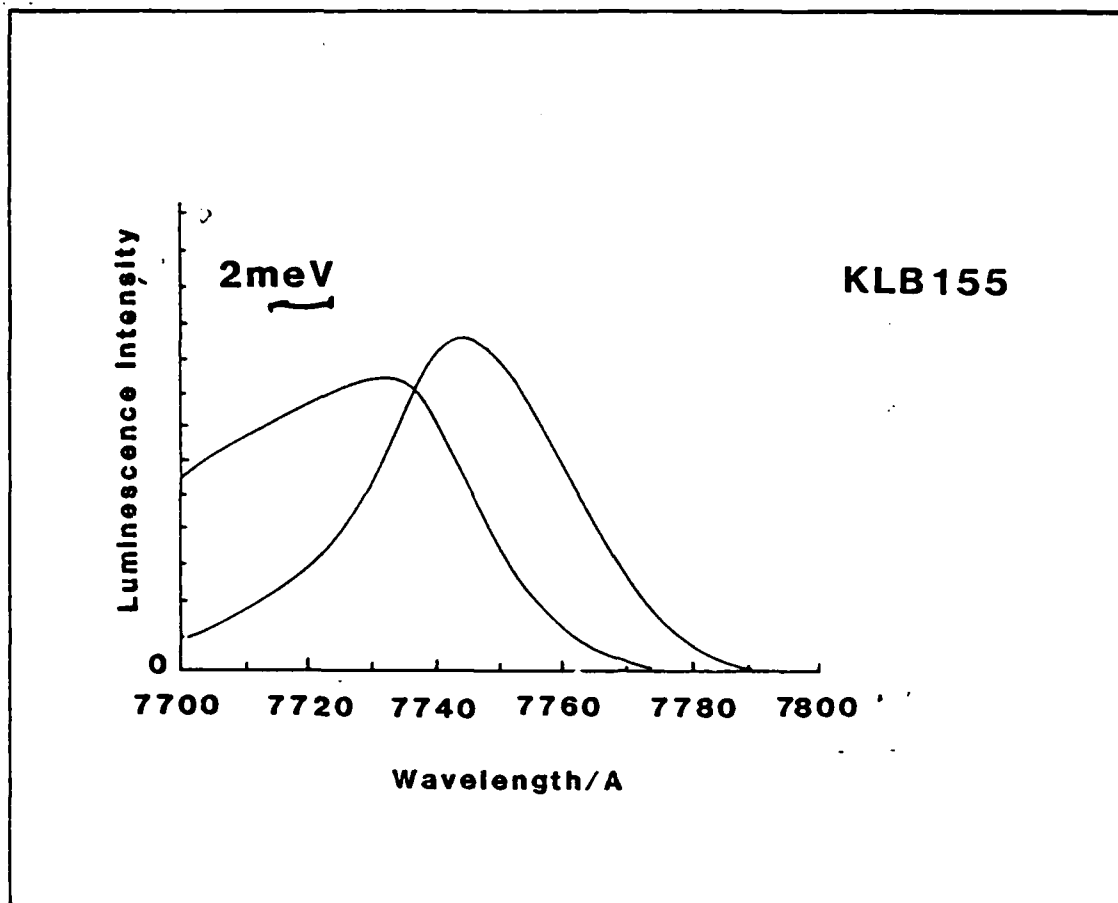


Figure 2

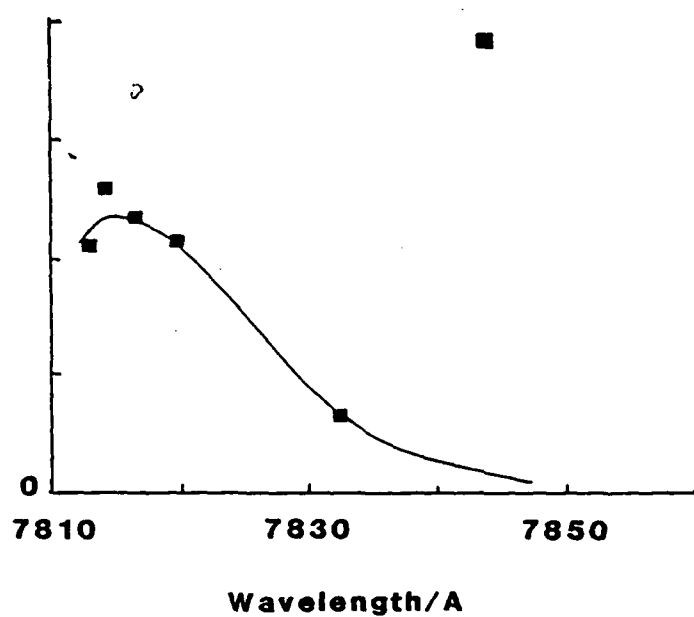


Figure 3 (a)

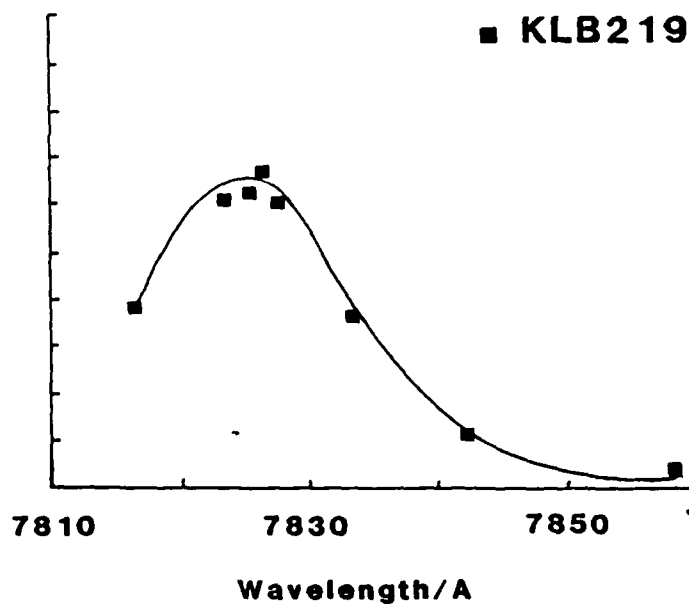


Figure 3 (b)

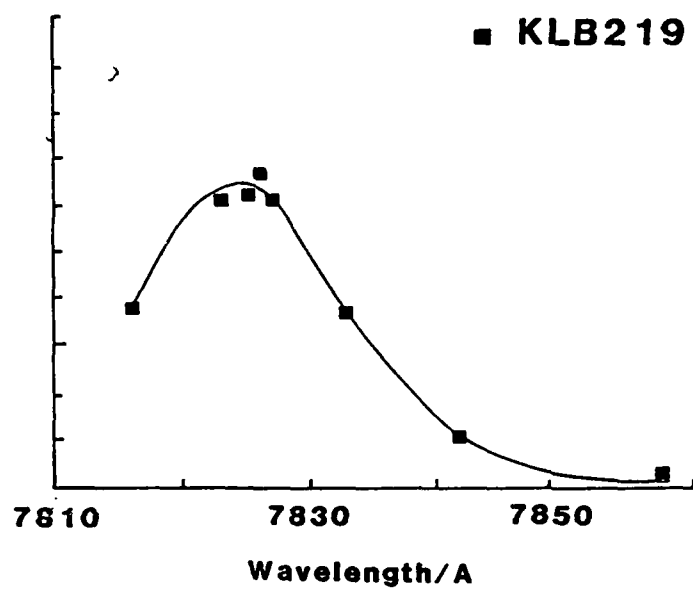


Figure 3 (b)

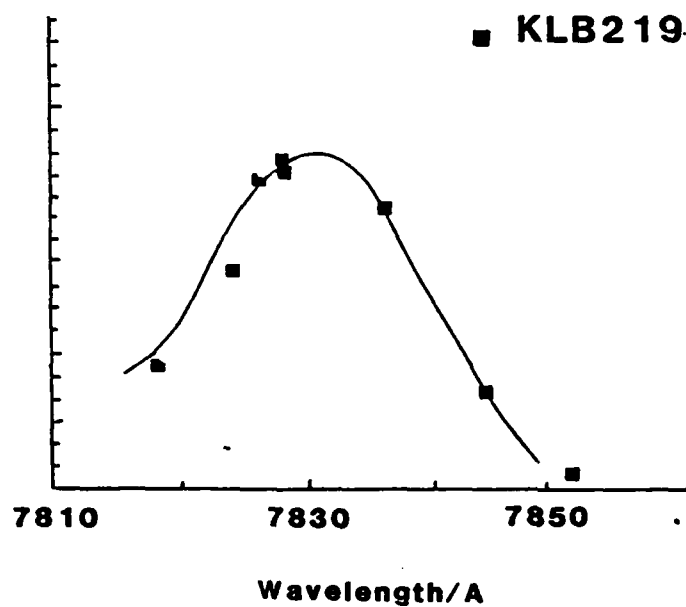


Figure 3 (c)

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DATE: 15 February 1988

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