Photoluminescence assessment of B, P, and Al in Si wafers: The problem of sample heating by a laser beam

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A line shape analysis of free-exciton low-temperature (4.2 K) photoluminescence spectra is applied to monitor the heating of Si samples due to an exciting Ar^+-ion laser beam. The heating is studied in dependence on the laser beam intensity. The temperature as a line shape fitting parameter can be established within ±0.15 K. Continuous minor increase of the sample temperature is observed up to an intensity of ~ 200 W cm^{-2}, followed by an abrupt temperature jump up to tens of K. Possible inaccuracies, resulting from the heating, in photoluminescent quantitative determination of boron, phosphorus, and aluminum are discussed.

I. INTRODUCTION

Characterization of silicon wafers by means of low-temperature photoluminescence measurements has been applied for more then 10 years. Tajima[1,2] proposed a method for determining quantitatively the contents of boron and phosphorus in lightly doped Si crystals, based on a properly calibrated ratio of intensities of bound exciton (BE) to free exciton (FE) photoluminescent lines. The method is applicable in the impurity range of $\sim 10^{11}$–10^{15} cm^{-3}. Similar calibration curves were later reported by other authors[3–6] and the method has also been applied to aluminum and other shallow donors/acceptors in silicon.[5,7,8] It is worth mentioning also that nowadays the purity of II-VI semiconductors (CdTe,[9] ZnSe,[10]) is being increasingly studied by a similar quantitative photoluminescent manner. However, in order to ensure sufficient accuracy and reproducibility of dopant concentration measurements by means of photoluminescence spectra, rather severe experimental conditions have to be assured. This is especially important for the sample temperature, because the ratio of BE to FE lines is strongly temperature dependent[11] due to the low exciton localization energy on shallow donors and acceptors (of the order of several meV[12]): the raise of the sample temperature causes the release of excitons from the shallow traps and therefore FE luminescence intensity grows with increasing temperature at the expense of the BE intensity. A quite natural way to maintain a fixed temperature of the samples is to immerse them in liquid helium at the boiling point $T=4.2$ K. However, possible local heating of the sample by the exciting laser beam [the use of 488 or 514 nm lines from a continuous wave (cw) Ar^+ laser is the most common for this purpose] remains as an open problem. This problem of uncontrolled sample heating by a fraction of the excitation energy dissipated nonradiatively in the crystal is of general nature in laser-excited photoluminescence spectroscopy of semiconductors. In silicon, however, like in other indirect-gap semiconductors, the broadening of the FE emission line can be used to study the sample temperature in a quantitative manner. This broadening reflects kinetic energy distribution in the FE gas and can be therefore described by a simple analytic expression—a Maxwell–Boltzmann distribution function. The temperature of the excitonic gas enters explicitly into the line shape formula and temperature can thus be established in this way noncontactly, by optical means.

In this article we report a detailed study of silicon samples heating by a cw Ar^+-ion laser. A line shape fitting procedure of the FE emission line, which includes both the Gaussian broadening due to phonons and the spectrometer slit broadening, is used for this purpose. The article is organized as follows. In Sec. II, we give a theoretical background on the FE line shape broadening and on the line shape fitting procedure. Section III presents experimental photoluminescence spectra of Si wafers at the bath temperature 4.2 K. The fitting is applied on them and the sample temperature increase is evaluated. Potential influence of the samples heating upon accuracy of quantitative determination of P, B, and Al in Si wafers is discussed in Sec. IV.

II. FREE-EXCITON EMISSION LINE SHAPE

A. FE line shape broadening

The participation of phonons in radiative recombination of FE in an indirect-gap semiconductor like Si makes it possible to describe the corresponding luminescence line shape by a simple relation:[13]

$$I_{\mathrm{FE}}(h\nu) \simeq \sqrt{\overline{h\nu}}\exp\left(-\frac{\overline{h\nu}}{k_B T}\right), \tag{1}$$

often called the Maxwell–Boltzmann distribution. Here, $\overline{h\nu} = h\nu - E_0$ is a kinetic energy of exciton translation motion and $E_0=E_g-E_x-h\Omega$ is a low-energy cutoff of the FE emission line. E_g, E_x, and $h\Omega$ stand for the energy gap, exciton binding energy, and energy of a phonon emitted in the recombination act, respectively. The k_BT has its usual meaning of a temperature Boltzmann factor.

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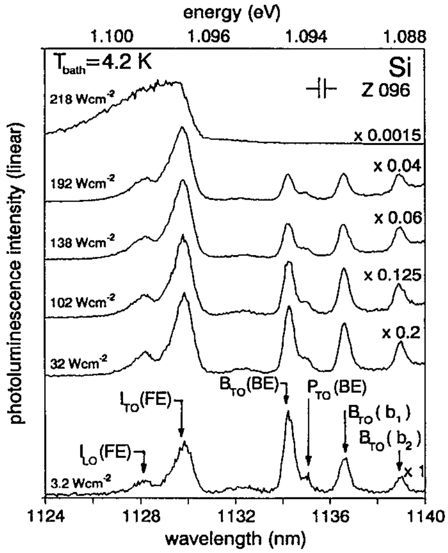

FIG. 1. Photoluminescence spectra of the Si sample Z 096 obtained with increasing excitation intensity (indicated at each spectrum on the left). Bath temperature $T=4.2$ K.

In silicon, symmetry selection rules allow for all types of phonons [i.e., LA (longitudinal-acoustic), TA (transverse-acoustic), LO (longitudinal-optic), and TO (transverse-optical)] to participate in FE radiative recombination. The most intense luminescence lines are, however, those involving TO and LO phonons. Due to a small difference between $\hbar\Omega_{TO}$ and $\hbar\Omega_{LO}$ ($\Delta_{LO/TO}=\hbar\Omega_{TO}-\hbar\Omega_{LO}=1.8$ meV[14]), these two lines denoted usually I_{TO}(FE) and I_{LO}(FE), are partly overlapping in the photoluminescence spectrum (see, e.g., Fig. 1). Our considerations throughout the article will be limited to this "doublet" $I_{TO}(FE)/I_{LO}(FE)$. These lines are also being generally applied to the quantitative photoluminescent determination of shallow donor and/or acceptor concentration in Si.[1-8]

It follows from Eq. (1) that the full width at half maximum (FWHM) of the FE emission line δ grows linearly with increasing temperature,

$$\delta \simeq 1.8\ k_BT. \tag{2}$$

Physically it means a growth of the mean kinetic energy of the FE gas with increasing T. The principal possibility of using Eq. (2) to measure optically the temperature of a luminescing sample is thus immediately seen, assuming Eq. (1) is of sufficient accuracy to describe the real FE line shape. However, this is not the case. There are two mechanisms contributing, apart from the temperature, to the broadening of the FE line. These are so-called Gaussian broadening and a spectrometer slit broadening.

The origin of the Gaussian broadening is supposed to be due to phonon collisions. It can be described as[15]

$$G(E'-\overline{h\nu},\sigma)\simeq\frac{1}{\sqrt{2\pi}\sigma}\exp\left(-\frac{(E'-\overline{h\nu})^2}{2\sigma^2}\right), \tag{3}$$

where σ means the Gaussian linewidth in points of inflexion. This parameter can be simply related to the Gaussian FWHM $\sigma_G=2\sqrt{2\ln 2}\sigma$.

The spectrometer slit-broadening effect depends on the widths of entrance and exit slits of the spectrometer. In the usual case of equal widths, it has the form of an isosceles triangle with surface area normalized to unity

$$A(E-E',\Gamma)=(1/\Gamma)(1-|E-E'|/\Gamma)$$
$$\text{for}\quad E\in\langle E'-\Gamma,E'+\Gamma\rangle \tag{4}$$
$$A(E-E',\Gamma)=0\quad\text{elsewhere.}$$

The parameter Γ determines the slit width in energy units. However, owing to diffraction effects originating on the slit, better approximation of the slit-broadening function is again a Gaussian:[16]

$$A_G(E-E',\Gamma)\simeq\exp\left(-\frac{4\ln 2(E-E')^2}{\Gamma^2}\right), \tag{5}$$

where Γ can be determined experimentally as the FWHM of a spectral image of a narrow spectral line $\Delta(h\nu)\ll\Gamma$. The resulting FE line shape at the exit of the spectrometer is finally given by a convolution of Eq. (1) with Eqs. (3) and (5) as

$$I_{FE}^{\xi}(E)\simeq\int\left[\int\sqrt{\overline{h\nu}}\exp\left(-\frac{\overline{h\nu}}{k_BT}\right)\times\exp\left(-\frac{(E'-\overline{h\nu})^2}{2\sigma^2}\right)d(\overline{h\nu})\right]\times\exp\left(-\frac{4\ln 2(E-E')^2}{\Gamma^2}\right)dE'. \tag{6}$$

The index ξ denotes the individual phonon replicas. This relation is used throughout the article to fit the TO and LO replicas of the FE emission line.

B. The fitting procedure

We use the simplex method[17] to fit the experimental FE line shape to the theoretical formula (6). Due to the overlapping of the TO and LO phonon replicas, the resulting FE emission line shape can be written as

$$I_{FE}(E,E_0^{TO},\Delta_{LO/TO})\simeq I_{FE}^{TO}(E,E_0^{TO})+X_{LO/TO}I_{FE}^{LO}(E,E_0^{TO}+\Delta_{LO/TO}), \tag{7}$$

where $E_0^{TO}=E_g-E_x-\hbar\Omega_{TO}$ is the low-energy threshold of the TO-phonon replica and $X_{LO/TO}$ gives the ratio of heights of the LO and TO replicas, which is faintly temperature dependent. There are three fitting parameters: the temperature T (as an input estimate we take $T=4.2$ K, the bath temperature), the ratio $X_{LO/TO}$, and the threshold E_0^{TO} (for both $X_{LO/TO}$ and E_0^{TO}, the input estimates are taken from our experimental results). We should like to stress that we fit, in fact, the FE gas temperature, but this temperature is believed to be equal to the lattice temperature owing to a sufficiently long FE lifetime ($\geqslant 1$ μs) in silicon.

TABLE I. Electrical characteristics of the samples. N_A and N_D are dopant concentrations of acceptors and donors, respectively, ρ is the resistivity of the sample.

Sample	Type	ρ (Ω cm)	N_A (cm^{-3})	N_D (cm^{-3})
Z 096	FZ-*p*	2860.0	1.0×10^{13}	5.5×10^{12}
Z 106	FZ-*n*	1160.0	2.8×10^{12}	6.4×10^{12}
S3123	CZ-*p*	0.84	$>10^{16}$	$\approx5.0\times10^{15}$

Every iteration step being accomplished, a comparison of a deviation S of the theoretical line shape (7) from the experimental spectrum I_{exp} is made using the least-squares method:

$$S=\sum_{i=1}^{n} |I_{\text{exp}}(E_i)-I_{\text{FE}}(E_i,E_0^{\text{TO}},\Delta_{\text{LO/TO}})|^2. \tag{8}$$

The summation in Eq. (8) is performed over the whole set of $n=50\div60$ experimental points used in the minimization procedure. After every iteration, we also calculate the difference between two last successive values of S. If the difference is smaller than a predetermined value, the iteration process is finished.

III. EXPERIMENTAL RESULTS AND THEIR ANALYSIS

A. Experiment

Silicon samples, with dimensions of $\sim9\times5\times1$ mm^3, cut from dislocation-free wafers, were immersed in liquid helium (bath temperature $T=4.2$ K) and excited with a 488 nm line of a cw Ar$^+$ ion laser. The laser beam was focused onto the samples with a $f=5$ cm lens, the maximum excitation intensity being $I_m\sim700$ W cm^{-2}. The absolute value of the excitation intensity on the sample surface was established independently in two ways. First, it was calculated as a ratio of the measured laser power to the exciting beam waist cross section, taking into account attenuation in windows of the cryostat and in the liquid He. Second, the value of I_m was estimated from the measured threshold intensity for the onset of the electron-hole liquid photoluminescence band.[18] Both the values of I_m agree within $\sim30\%$.

Photoluminescence was detected in the backscattering geometry and dispersed by a double-grating $f=0.6$ m monochromator equipped with two 1200 grooves/mm gratings blazed at 1.2 μm. A liquid-nitrogen-cooled S1-type photomultiplier followed by a photon-counting detection system were used to record the photoluminescence spectra. The detection system was driven by a personal computer. The measured photoluminescence spectra were corrected for detection channel spectral response, which was calibrated with a tungsten strip lamp.

Two float-zone (FZ)-grown Si samples were mainly used in this study, one of *n*-type (Z 106) and the other of *p*-type (Z 096). Auxiliary experiments were performed on a Czochralski (CZ) *p*-type sample (S 3123). The basic characteristics of the samples are given in Table I.

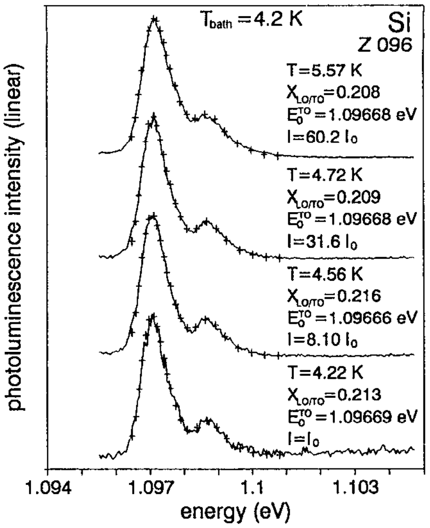

FIG. 2. Excitation intensity dependence of photoluminescence spectra for the Si sample Z 096 in the free exciton region. Bath temperature $T=4.2$ K. Theoretical fits of the LO and TO phonon replicas [Eq. (7)] are denoted by crosses, fitting parameters are indicated above each curve. Excitation intensity $I_0=3.2$ W cm^{-2}, Gaussian parameter $\sigma=0.22$ meV, spectrometer slit width 0.25 meV. The spectra are normalized.

B. Results of the fitting procedure

The overall view of the photoluminescence spectra of the Z 096 sample is shown in Fig. 1, the excitation intensity I being a parameter. With increasing I, the comparative predominance of FE features over the boron and phosphorus BE lines is clearly seen. (Sudden total disappearance of the extrinsic BE emission displayed at the uppermost spectrum in Fig. 1 will be discussed later on.) This effect is first of all due to the saturation of the BE photoluminescence.[5] However, sample heating can lead to the same observation, owing to the thermal release of the BEs. We apply, therefore, the fitting procedure to extract the sample temperature. Portions from the Fig. 1, containing the $I_{\text{TO}}(\text{FE})/I_{\text{LO}}(\text{FE})$ lines are shown enlarged in Fig. 2 along with the theoretical fitted line shapes (crosses) according to the Eqs. (6) and (7). Similar spectra for the sample Z 106 are presented in Fig. 3.

To obtain good fits, it turned out to be necessary to subtract background luminescence, which is slightly increasing in intensity towards the long-wavelength side of the FE emission (see, e.g., the lowermost spectrum in Fig. 1). It is also worth mentioning that, in order to obtain good fits as shown in Figs. 2 and 3, we have to introduce sample dependent values of $\Delta_{\text{LO/TO}}$ into Eq. (7): we take $\Delta_{\text{LO/TO}}=1.7$ meV for Z 096 and $\Delta_{\text{LO/TO}}=1.67$ meV for Z 106, while presently the generally accepted value of $\Delta_{\text{LO/TO}}$ is 1.8 meV.[14] Taking into consideration that LO and TO phonon energies were measured by optical modulation spectroscopy within absolute accuracy of $\eta=\pm0.4$ meV,[19] we may estimate the attainable accuracy of $\Delta_{\text{LO/TO}}$ to be $\Delta_{\text{LO/TO}}=[1.8\pm\sqrt{(2\eta^2)}]$ meV$=(1.8\pm0.56)$ meV. Both our values of $\Delta_{\text{LO/TO}}$ lie, therefore, well within this limit.

The fitting routine is then the following. For each sample, the spectrum measured with the lowest excitation in-

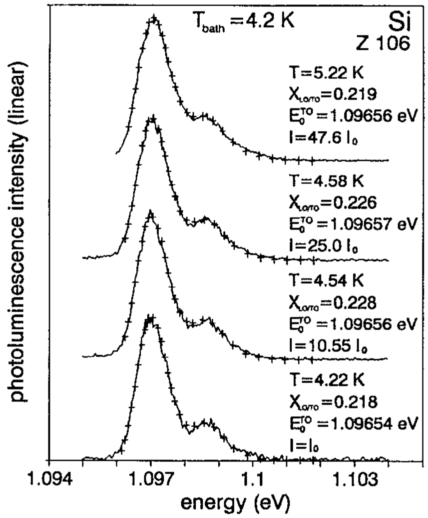

FIG. 3. Excitation intensity dependence of photoluminescence spectra for the Si sample Z 106 in the free exciton region. Bath temperature $T=4.2$ K. Theoretical fits of the LO and TO phonon replicas [Eq. (7)] are denotes by crosses, fitting parameters are indicated above each curve. Excitation intensity $I_0=4.0$ W cm^{-2}, $\sigma=0.33$ meV, spectrometer slit width 0.25 meV. The spectra are normalized.

tensity is fitted at first, with an assumption of negligible heating and thus fixed sample temperature $T=4.2$ K. In other words, we leave here the parameter σ to vary instead of the temperature. The obtained values of σ are subsequently used for higher excitation intensities as a fixed (sample dependent[20,21] but temperature independent) parameter to fit T, $X_{\mathrm{LO/TO}}$, and E_0^{TO}.

Resulting dependence of the sample temperature on the exciting intensity is shown in Fig. 4. Continuous temperature increase up to ~ 5.5 K for $I \sim 200$ W cm^{-2} is clearly seen, starting even at the lowest excitation intensity. Of course, a crucial point in this respect is a reliable estimate of accuracy of the temperature fits. We used a simple procedure to estimate this accuracy, namely, we calculated a series of theoretical line shapes differing step by step ΔT in temperature from the corresponding optimal fit obtained by the simplex method. Deviation of the theoretical curves from the experimental spectrum, as judged by the naked eye, became evident already at $\Delta T = \pm 0.15$ K. Therefore, we put this value of ΔT as an upper limit of the accuracy of the temperature determination.

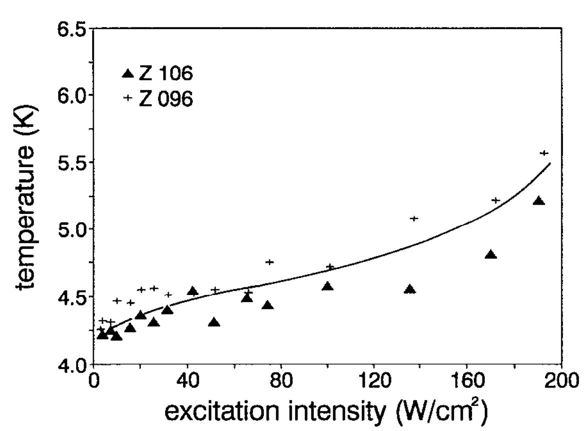

FIG. 4. Dependence of the local temperature of the Si samples Z 096 and Z 106 on the excitation intensity. Bath temperature $T=4.2$ K. The continuous line is drawn as a guide for the eyes.

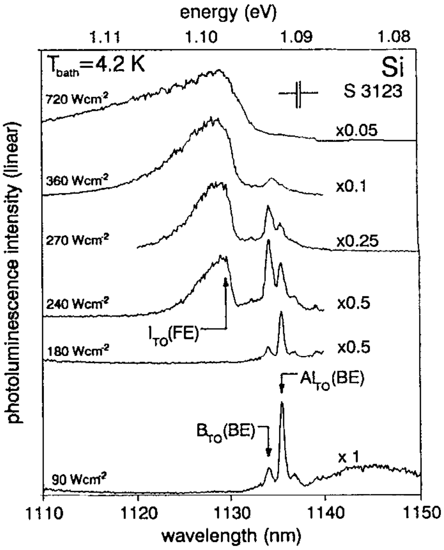

FIG. 5. Photoluminescence spectra of the sample S 3123. Dominant impurities are Al ($>10^{16}$ cm^{-3}) and As ($\approx 5.10^{15}$ cm^{-3}). The line As$_{\mathrm{TO}}$(BE) is not resolved from Al$_{\mathrm{TO}}$(BE), the line B$_{\mathrm{TO}}$(BE) originates from the residual boron impurity ($\approx 10^{13}$ cm^{-3}). Excitation intensity I is indicated above each spectrum. The appearance of a wide free exciton emission band for $I > 220$ W cm^{-2} can be correlated with the uppermost spectrum in Fig. 1.

When further increasing the excitation intensity of the laser beam, dramatic qualitative changes can be observed in the photoluminescence spectra as shown in Fig. 5. This figure represents emission spectra of the comparatively highly Al- and As-doped sample S 3123. Because of high impurity concentration, no FE luminescence can be seen up to $I \sim 180$ W cm^{-2}. For $I \sim 220$ W cm^{-2}, an abrupt appearance of the I_{TO}(FE) line accompanied by a noticeable change in the B$_{\mathrm{TO}}$/Al$_{\mathrm{TO}}$ intensity ratio occurs. We believe this is due to high local sample heating, probably caused by the loss of thermal contact with the bath. The sample temperature raises above the critical temperature T_c of the electron-hole liquid (EHL) $T_c = (25 \pm 5)$ K.[22] Evaporation of the liquid implies a sudden increase of the FE gas density and also an increase of the number of excitons captured at impurity centers. Experimentally it means the growth of both FE and BE emission intensity. Further increase of excitation intensity leads gradually to the thermal dissociation of BE complexes and finally only the FE line persists, owing to sufficiently high FE binding energy (14.7 meV) (see the uppermost spectrum in Fig. 5). This conjecture is fully supported by our estimates of the temperature when applying Eq. (2) to the very broad FE lines in Fig. 5. (We do not apply the fitting procedure here to establish T because of possible uncertainty originating in excessive spectral widths and *full* superposition of both phonon replicas in the FE line.) Temperatures in the range 24 K$\leqslant T \leqslant$75 K are obtained, i.e., values altogether higher than T_c (see Table II). Moreover, clear disappearance of the wide EHL emission peaked at ~ 1145 nm un-

TABLE II. Dependence of the Si sample (S 3123) temperature T on excitation intensity I as estimated by Eq. (2) from Fig. 5.

I (W cm^{-2})	T (K)
240	24
270	31
360	42
720	75

der excitation levels $I \geqslant 220$ W cm^{-2} is a direct evidence of the thermal dissociation of the EHL (Fig. 5). An overall view of the temperature against I on the basis of combination of the data obtained on samples Z 096 and S 3123 is shown in Fig. 6.

IV. DISCUSSION

It follows now from our results that the reduction of BE/FE intensity ratio with increasing I in Fig. 1 is not caused by heating, because, according to Fig. 4, the maximum temperature reached in this case is $T \sim 5.6$ K. At temperatures below ~7 K, evaporation of bound excitons from B, P, and Al centers is negligible compared to decay[12] as the binding energies of BE on boron, aluminum, and phosphorus are relatively high in this respect: $\varphi_B = 3.6$ meV, $\varphi_{Al} = 4.4$ meV, and $\varphi_P = 4.9$ meV.[12] Therefore, we conclude that the heating up to 5.6 K by the excitation radiation $I < 200$ W cm^{-2} is not sufficient to release the excitons and that the changes in photoluminescence intensity like in Fig. 1 are almost solely due to the saturation of impurity centers. On the other hand, certain precautions may be judicious, even when dealing with Fourier transform photoluminescence spectroscopy as described by McColley and Lightowlers.[5] Though the authors do not give the excitation intensity used, it can be estimated from their data to be rather high, of the order of ~100 W cm^{-2}. The dimensions of the samples being comparable to ours (and being typical for photoluminescence assessment of silicon in general), we estimate on the basis of our results the temperature of luminescence emitting region of samples to be $T \sim 4.7$ K. Heating is not critical, certainly, but at the same time it is perhaps not fully correct to state that the temperature is maintained very close to 4.2 K. In any case, the use of a bath LHe cryostat is indispensable, otherwise a temperature rise would be uncontrollable.

In conclusion, applying the method of contactless temperature determination by FE photoluminescence line shape fitting, we have shown that heating of Si samples by an Ar$^+$-ion laser beam is measurable. The temperature of the laser-excited spot amounts up to $\sim 5.7 \pm 0.15$ K at $I \approx 200$ W cm^{-2} if the sample is immersed in liquid He at atmospheric pressure. At excitation levels usually used in the photoluminescence analysis of Si, which are typically at least several times lower, the heating does not play any significant role and its influence upon results of quantitative analysis of B, Al, and P dopants may be considered negligible within an analysis accuracy attainable at present.[5,8] Care has to be taken, however, not to excite excessively and to avoid the loss of thermal contact with the helium bath.

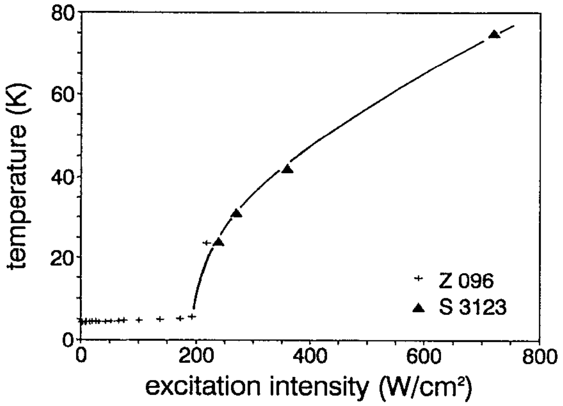

FIG. 6. Dependence of the local temperature on the excitation intensity for the samples Z 096 (below 200 W cm^{-2}) and S 3123 (above 200 W cm^{-2}). Bath temperature $T = 4.2$ K. The continuous line is drawn as a guide for the eyes.

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