

Influence of exciton impact ionization and illumination intensity on the exciton-polariton reflectance of GaAs

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External electric fields cause impact ionization of excitons in GaAs. We investigated the influence of impact ionization on exciton-polariton reflectance spectra. The change in the reflectance spectra due to the external field is similar to the change obtained by reducing the illumination intensity to very small values. This joint behavior is explained in terms of band bending near the crystal surface and related damping of the exciton polariton.

I. INTRODUCTION

The reflectance of exciton polaritons in semiconductors has been the subject of extensive studies in recent years. These studies investigated basic physical properties of exciton polaritons and dealt with the influence of external perturbations on the exciton polariton, especially with the influence of electric fields applied perpendicularly to the crystal surface,^{1,2} magnetic fields,³ electron-beam irradiation,^{4,5} or surface treatment.⁶⁻⁸

The experiments on the exciton-polariton reflectance indicated that strong internal electric fields, caused by band bending near the crystal surface, evoke a field-induced ionization of excitons.⁹ The field strengths necessary to *field ionize* excitons are in the order of several kV/cm.¹⁰ A surface layer is created where excitons cannot exist. Experimentally, the influence of this internal field has been studied, for example, by applying additional external electric fields *perpendicularly* to the sample surface^{1,2} or by altering the illumination intensity of the incident light.¹¹ In both cases the external perturbation modifies the band bending near the crystal surface, alters the internal field, and thus changes the width of the exciton-free surface layer.

Much lower electric fields in the order of a few V/cm are sufficient to *impact ionize* excitons.¹²⁻¹⁴ Free charge carriers are accelerated in an electric field applied *parallel* to the sample surface and gather kinetic energy sufficient to dissociate free excitons via impact. This mechanism can be expected to act directly on the damping of the exciton polariton. Impact ionization of excitons thus should change the reflectance spectrum in a manner describable by a field-dependent damping constant.

The aim of this paper is to study the behavior of the exciton-polariton reflectance line shape

under impact-ionization conditions. We measure the reflectance of *n*-GaAs in an electric field applied parallel to the crystal surface and compare the results with those obtained by varying the illumination intensity. It turns out that the reflectance spectra change under impact-ionization conditions in the way they do when the illumination intensity is reduced to a very small value.

We describe this experimental behavior by a theoretical model which uses a dielectric function with a spatially varying damping constant.^{15,16} Both impact-ionization and illumination-intensity alteration act on this damping constant. The acting mechanism, though, differs for the two cases.

We check our interpretation by measuring the lifetime of the free exciton in a time-resolved luminescence experiment. We can identify an impact-ionization-induced change of the exciton damping by the corresponding change of the exciton lifetime.

II. EXPERIMENTAL

The GaAs samples used in our experiments were layers grown by liquid-phase epitaxy on semi-insulating substrates. They exhibited *n*-type conductivity with room-temperature carrier concentrations between 5×10^{13} and 1×10^{14} cm⁻³. The mobilities at 77 K ranged between 115 000 and 130 000 cm²/Vsec. The electric field was applied parallel to the sample surface by means of two Ohmic contacts. The field strengths quoted in this paper are the quotients of the voltages applied and the contact spacings. The fields may not be correct in absolute units, as the influence of the photocreated carriers on the conductivity in the illuminated spot is neglected. But, the way in which the field strengths are computed is consistent for all measurements. The samples were

held at 1.8 K in a helium-immersion cryostat.

We used nonmonochromatic light from a tungsten-iodine lamp for the reflectance measurements under impact-ionization conditions. The power density of the incident light beam was about 10 mW/cm^2 . The reflected intensity was detected with a 0.75-m grating monochromator, and the wavelength resolution was about 0.15 \AA .

A different experimental setup was used for the study of the illumination-intensity dependence. In these experiments, the sample was illuminated by an argon-laser beam ($\lambda = 488 \text{ nm}$) with an extremely low intensity $I_L \approx 130 \text{ nW/cm}^2$ and multiples thereof. Simultaneously we irradiated the sample with monochromatic probe light (line-width about 0.2 \AA). This light was obtained from a tungsten-iodine lamp and a 1-m grating monochromator and had an intensity $I_p \approx 150 \text{ nW/cm}^2$. We measured the reflected intensity of the probe beam as a function of energy under different laser-illumination intensities I_L .

We also varied the probe-beam intensity I_p instead of the intensity I_L of the additional laser beam and obtained the same results. However, such spectra with different probe-beam intensities are not so easily comparable because of different intensity scales and different time constants of the amplifier.

In all experiments, the spectra were recorded with a single-photon counting system providing excellent signal-to-noise ratios even at low excitation powers and consequently low reflected intensities.

The time-resolved measurements of the exciton luminescence were performed using the technique of delayed coincidence.¹⁷ A mode-locked argon laser ($\lambda = 514.5 \text{ nm}$) was used as excitation source. It provided light pulses of approximately 0.3-nsec pulse width at a repetition frequency of 95.3 MHz. The time resolution of the detection system was about 0.7 nsec.

III. RESULTS

Reflectance spectra of one of our GaAs epitaxial layers in the energy region of the free-exciton polariton are shown in Fig. 1. The lowest spectrum of Fig. 1 was taken without an external electric field F . The reflectance spectra are changed if an external electric field with field strength F greater than the threshold field for impact ionization of excitons ($F \approx 1 \text{ V/cm}$ in this sample) is applied parallel to the surface (see upper spectra of Fig. 1). A second minimum in reflectivity appears near the longitudinal resonance energy $\hbar\omega_L = 1.51515 \text{ eV}$. This minimum shifts to lower energies with increasing field

strength while the maximum of reflectivity decreases. The spectrum in Fig. 1 at $F = 1.52 \text{ V/cm}$ demonstrates that even a second maximum arises at the longitudinal resonance energy under increased field strengths.

The reflectance spectra of Fig. 1 with different impact-ionization fields resemble spectra taken by Fischbach *et al.*,¹¹ who varied the illumination intensity. We repeated the experiments of Fischbach *et al.* using the sample of Fig. 1. The variation of the illumination intensity was achieved by an additional laser beam as described above.

Figure 2 displays the reflectance spectra obtained in these experiments. The lowest spectrum was measured at high illumination intensity ($I = I_p + 250I_L$). Its line shape equals that of the spectrum with $F = 0 \text{ V/cm}$ in Fig. 1. Such a reflectance spectrum does not change with illumination intensity I_L as long as I_L stays higher than about 40 \mu W/cm^2 . A decrease of I_L below this threshold value, however, causes remarkable changes in the reflectance spectra as can be seen in the upper part of Fig. 2. These changes demonstrate how careful one has to be in interpreting reflectance spectra at very low illumination intensities.¹⁸

The similar behavior of the reflectance line shape under impact ionization and illumination reduction (compare Figs. 1 and 2) implies that a simultaneous variation of the two different experimental parameters amplifies the changes in the

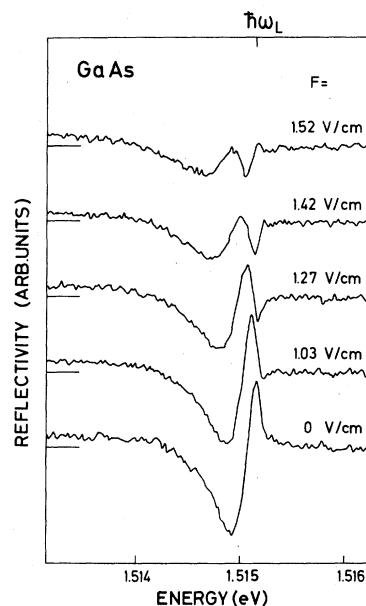


FIG. 1. Experimental reflectance spectra of GaAs vs energy in the exciton energy region for various field strengths F parallel to the crystal surface. The horizontal lines assign equal reflectivities.

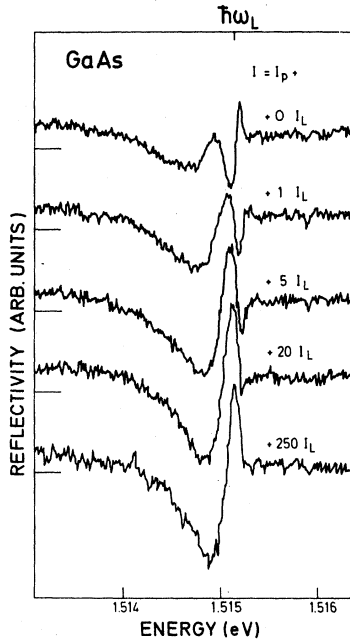


FIG. 2. Experimental reflectance spectra of GaAs vs energy for various illumination intensities I . I_p denotes the intensity of the probe beam, I_L is the intensity unit of the additional laser beam. The horizontal lines assign equal reflectivities.

reflectance spectra. We consequently combined the two experimental techniques using the intensity $I = I_p + 5I_L$ in Fig. 2 and applied an additional impact-ionization field. The result is seen in Fig. 3. These reflectance spectra suggest that changes of the impact-ionization field and of the illumination intensity affect the reflectance spectra additively and in the same manner.

IV. DISCUSSION

The most significant change of the reflectance spectra in Figs. 1–3 is the appearance of a second minimum and a second maximum of the reflectivity near the longitudinal resonance frequency ω_L . An explanation of the experimental results has to reproduce this significant feature.

Initially we followed the standard procedure and calculated reflectance spectra of a crystal whose dielectric function ϵ for excitons shows spatial dispersion for given wave vector \vec{k} and frequency ω (Ref. 9)

$$\epsilon(\omega, k) = \epsilon_\infty \left(1 + \frac{\omega_L^2 - \omega_T^2}{\omega_T^2 - \omega^2 + \beta k^2 - i\omega\Gamma} \right). \quad (1)$$

ϵ_∞ is a frequency- and wave-vector-independent background dielectric constant ($\epsilon = \epsilon_\infty$ for $\omega \rightarrow \infty$), ω_T and ω_L the transverse and longitudinal resonance frequencies at $k=0$, Γ the empirical damp-

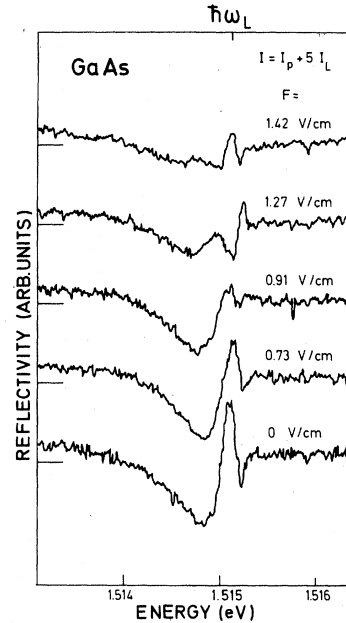


FIG. 3. Experimental reflectance spectra of GaAs vs energy for the illumination intensity I . The field strength F of an impact-ionization field is varied. The horizontal lines assign equal reflectivities.

ing constant, and $i = \sqrt{-1}$. The influence of spatial dispersion is described by $\beta k^2 = (\hbar\omega_T/M)k^2$, where M is the effective exciton mass and \hbar is Planck's constant divided by 2π .

We included in our calculations an exciton-free surface layer as proposed by Hopfield and Thomas.⁹ They assume a model with a step in the dielectric function: an exciton-free layer of dielectric constant ϵ_∞ on top of the bulk of the crystal, the latter being described by the dielectric function $\epsilon(\omega, k)$.

We were able to fit the lowest spectra of Fig. 1 ($F=0$ V/cm) and of Fig. 2 ($I = I_p + 250I_L$) with this model using parameters given in the literature.¹⁹ However, we could not reproduce the characteristic features of the other spectra in Figs. 1 and 2 with any set of parameters—neither with spatial dispersion ($\beta \neq 0$) nor without spatial dispersion ($\beta = 0$).

To overcome this failure we improved our calculations by a model which uses a spatially varying continuous dielectric function $\epsilon(\omega, x)$ instead of a step in the dielectric function. In this model, the damping parameter Γ used in ϵ is assumed to decrease exponentially with distance x from the surface¹⁵:

$$\Gamma(x) = (\Gamma_s - \Gamma_B)e^{-x/A} + \Gamma_B. \quad (2)$$

Γ_s is the damping parameter at the surface ($x=0$), Γ_B the damping parameter in the crystal

bulk ($x \rightarrow \infty$), and A the spatial decay constant. The continuous function $\Gamma(x)$ is approximated by a step function of at least 100 steps for the numerical evaluation. This model does not include spatial dispersion (i.e., $\beta=0$ in the model), and its properties will be discussed in more detail elsewhere.¹⁶

First we fitted the lowest reflectance spectrum of Fig. 1 ($F=0$ V/cm) and of Fig. 2 ($I=I_p+250I_L$). The parameters obtained are listed in Table I. The calculated spectrum is shown in the lowest part of Fig. 4 ($A=190$ Å). It reproduces the experimental spectra very well.

An increase of the decay constant A in the exponential damping parameter $\Gamma(x)$ yields the spectra presented in the upper part of Fig. 4. These spectra exhibit the characteristic features of the experimental spectra of Figs. 1 and 2 and show especially the two minima and two maxima in reflectivity.

The most characteristic changes in the reflectance spectra occur at the longitudinal resonance frequency ω_L . Therefore, we plotted the experimental and theoretical reflectivities at $\hbar\omega_L$ of Figs. 1 and 4. The result is shown in Fig. 5: the reflectivity at $\hbar\omega_L$ versus the field strength F of impact ionization (left part of Fig. 5) and versus the decay constant A (right part of Fig. 5). The comparison of the experimental data and of the theoretical results substantiates that the model using a damping parameter with spatial dependence perpendicular to the surface reproduces the observed nonmonotonic behavior.

The following discussion pertains to the physical meaning of the described model. It is well known that an always-present surface-charge density causes a bending of the energy bands near the surface.²⁰ This band bending leads to a depletion of carriers for n -type material and results in an electric field near the surface. This electric field causes a field ionization of excitons, and the ionization may be described by a damping parameter $\Gamma(x)$ with a spatial dependence perpendicular to the surface. The band bending therefore justifies the use of a spatially dependent damping parameter.

An illumination of the crystal surface creates free charge carriers which compensate the sur-

TABLE I. Parameters of the free exciton in GaAs used for the calculations.

$\hbar\omega_T$ (eV)	1.51499
$\hbar\omega_L$ (eV)	1.51515
ϵ_∞	12.6 ^a
$\hbar\Gamma_s$ (meV)	8.0
$\hbar\Gamma_B$ (meV)	0.05

^aTaken from Ref. 19.

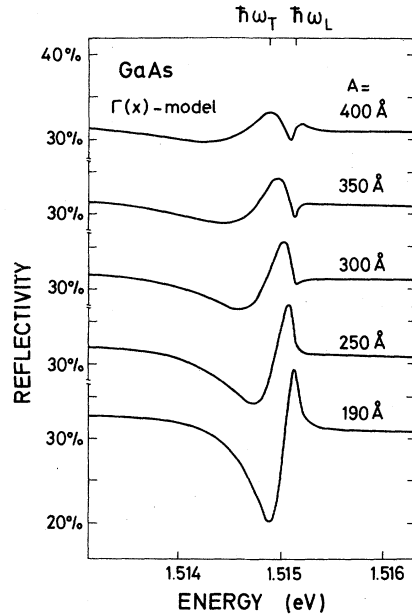


FIG. 4. Calculated reflectance spectra of GaAs vs energy for an exponential spatial dependence of the damping parameter $\Gamma(x)$. The spatial decay constant A is varied. The parameters used for the calculation are listed in Table I.

face space charge and reduce the band bending at the surface. Consequently, a reduction of the illumination intensity decreases the concentration of photocreated free charge carriers. This reduction effects an increase in thickness of the surface-space-charge region and so extends the ionizing field further into the bulk.¹⁸ Hence, the layer in which the exciton polariton is damped will be increased. This behavior may be described by a larger decay constant A in the damping parameter $\Gamma(x)$ (cf. Figs. 2 and 4).

Now we turn from field ionization to impact

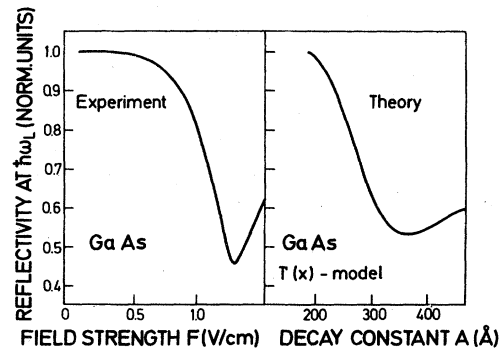


FIG. 5. Experimental reflectivity at the energy $\hbar\omega_L$ versus the field strength F of impact ionization (left-hand part) and theoretical reflectivity at $\hbar\omega_L$ versus the spatial decay constant A (right-hand part).

ionization. The rate of impact ionization is determined by the concentration of mobile ionizing carriers. This concentration is diminished near the surface because of the band bending. Thus the impact ionization and the corresponding additional damping of excitons is more efficient in the bulk than in the region near the surface. This behavior near the surface may be described by an increase of the decay constant A in $\Gamma(x)$, too (cf. Figs. 1 and 4). An additional increase of the damping parameter Γ_B in the crystal bulk has no essential influence on the reflectance spectra.

These considerations disclose that impact ionization of excitons by accelerated carriers and field ionization of excitons by band bending at the crystal surface cause similar and additive changes in the reflectance spectra. The superposition takes place via a superposition of damping by impact ionization and of damping by field ionization which both show a spatial dependence perpendicular to the surface (cf. Fig. 3).

The change of the decay constant A due to external perturbation has been obtained indirectly from the experiments. Measured spectra have been fitted by a theoretical model containing A as adjustable parameter. In the case of impact ionization, however, a change of the decay constant A and thus of the damping parameter Γ can be measured directly. The damping parameter Γ for excitons is proportional to the constant $C = 1/\tau$, which describes the exponential temporal decay rate of excitons of density N (τ is the exciton lifetime):

$$\frac{dN}{dt} = -CN + G, \quad (3)$$

where G is the generation rate of excitons.

G equals zero for a *time-resolved* luminescence experiment after the exciting laser pulse is switched off. Thus, from Eq. (3) it follows that

$$N = N_0 e^{-Ct}. \quad (4)$$

The time-resolved luminescence intensity I originating from the radiative decay of excitons is proportional to the actual concentration N of excitons (without impact-ionization field: $C = C_0$):

$$I \sim N = N_0 e^{-C_0 t}. \quad (5)$$

Impact ionization of excitons opens an additional nonradiative decay channel for free excitons described by an additional decay rate C_I . The overall decay rate C of excitons is then given by $C = C_0 + C_I$. An increase of exciton damping due to impact ionization is therefore expected to yield a decrease of the exciton lifetime $\tau = 1/C = 1/(C_0 + C_I)$.

Figure 6 shows the result of such a time-resolved luminescence experiment. The integral luminescence intensity of the free exciton is plotted as a function of the time elapsed after pulsed excitation. We measured a decay time τ_0 of 70 nsec at zero field. An additional impact-ionization field parallel to the surface reduces the radiative lifetime of the free exciton to $\tau = 3.5$ nsec corresponding to an additional nonradiative decay channel.

It is not possible to perform a time-resolved luminescence experiment in order to study the influence of illumination-intensity changes on the exciton lifetime. The luminescence is far too weak to be detectable at the necessary low excitation intensities.

Equation (3) can be used to describe *steady-state* luminescence, too. A steady-state luminescence experiment has a constant generation rate G of excitons; dN/dt equals zero. The luminescence intensity I_0 without impact ionization then follows from Eq. (3):

$$I_0 \sim N = G/C_0. \quad (6)$$

Impact ionization of excitons opens an additional nonradiative decay channel with a decay rate C_I ; the overall excitonic decay rate under impact ionization then is $C_0 + C_I$. We assume that the generation rate G has not changed essentially in the electric field. This assumption is reasonable because the excitonic polarizability and thus the

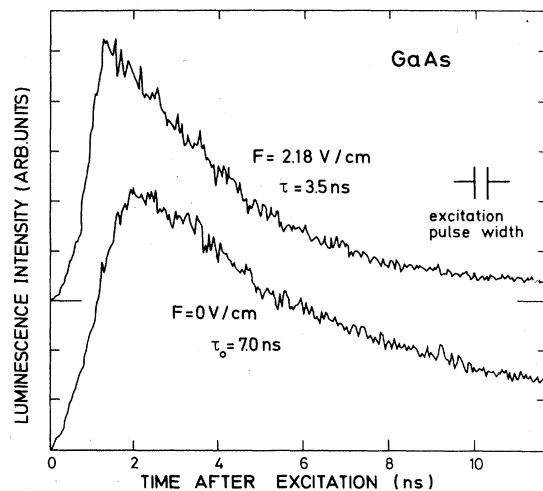


FIG. 6. Experimental time-resolved luminescence intensities of the free exciton in GaAs for different field strengths F parallel to the crystal surface. The upper value of F is not directly comparable to those of all other figures because this figure was taken under high laser excitation conditions. The temporal decay constant τ was obtained by fitting the experimental spectra with an exponential decay function.

longitudinal-transverse resonance-energy splitting would otherwise be changed essentially. Such a change, however, is not observed in the experiments. Therefore, the luminescence intensity I with impact ionization is given by

$$I/I_0 = 1/(1 + C_I/C_0). \quad (7)$$

Our model should be able to predict, at least qualitatively, the behavior of the luminescence intensity as a function of the electric field. We infer from our experiments that impact ionization increases the damping of exciton polaritons. This damping is described in the model by a spatially varying damping parameter Γ with decay constant A . The decay constant A depends on the applied electric-field strength.

The left-hand side of Fig. 7 shows the experimental steady-state luminescence intensity integrated over the free exciton energy region versus the field strength F of the impact-ionization field.²¹

The corresponding theoretical luminescence intensity of Eq. (7) was calculated as a function of the spatial decay constant A used for the damping parameter $\Gamma(x)$. The value of A for the best reflectance line-shape fitting ($A = 190 \text{ \AA}$, cf. Fig. 4) was used to calculate Γ for the case without impact ionization, where $\Gamma \sim C_0$. The decay constant A is greater than 190 \AA for increasing impact ionization, where Γ is proportional to $C_0 + C_I$. Using the value of Γ at a depth of, e.g., $x = 1000 \text{ \AA}$, the result for I/I_0 is shown on the right-hand part of Fig. 7. (The choice of $x = 1000 \text{ \AA}$ is arbitrary. Other values of x yield similar results.)

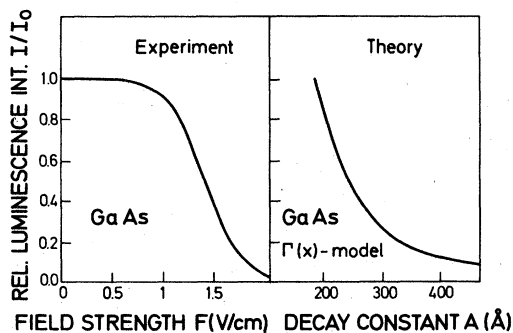


FIG. 7. Experimental steady-state luminescence intensity integrated over the free exciton energy region vs the field strength F of impact ionization (left-hand part) and theoretical luminescence intensity for the free exciton in GaAs vs the spatial decay constant A (right-hand part). I_0 denotes the luminescence intensity at $F = 0 \text{ V/cm}$, respectively $A = 190 \text{ \AA}$.

As can be seen, the theoretical luminescence intensity decreases with increasing spatial decay constant A .

The comparison of the experiments and the theoretical results in Fig. 7 exhibits that the model predicts the experimental results in a good qualitative way. This finding confirms the interpretation of our results.

V. CONCLUSIONS

The theoretical results reproduce the characteristic features of the experiments very well. We obtained a good quantitative agreement between theory and experiment, especially for the spectra at high illumination intensity and without impact ionization (lowest spectra of Figs. 1, 2, and 4). This agreement is not as satisfactory under low excitation and/or impact-ionization conditions (upper spectra of Figs. 1, 2, and 4). We suppose that the functional description of the damping parameter $\Gamma(x)$ and of the dielectric function ϵ is still incomplete.

The first problem is the inclusion of spatial dispersion of excitons. Spatial dispersion has not been included because of the difficulty in defining the additional boundary conditions at the internal interfaces in the described model.^{15,16}

The second question is whether the damping parameter has a spatial dependence described by a simple exponential function. The connection between our calculations and the explanation in terms of band bending at the surface is given by the relation between the damping parameter and an electric ionization field which decreases linearly from the surface into the crystal bulk.

Attempts to use a corresponding linear decrease of the damping parameter $\Gamma(x)$ gave no useful result. A constant surface-damping parameter up to a certain distance from the surface and a matching decrease to the bulk damping were not able to reproduce the characteristic experimental features either. The exponential decrease of the damping parameter was the only model with which we could describe the experimental results in an adequate way.

These considerations show that the described model with an exponential decrease of the damping parameter reproduces the experimental characteristics, but has to be regarded as a first attempt. The main remaining question is the correct connection between the damping parameter Γ used in the calculations and the electric fields near the crystal surface under the experimental conditions used in this work.

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