Elastic Scattering of Exciton Polaritons by Neutral Impurities

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A model based on the elastic scattering of exciton polaritons from residual neutral impurities is proposed to account for the line shape of the exciton-polariton photoluminescence emission in GaAs at low temperatures. The good agreement obtained with experimental spectra emphasizes the importance of impurity scattering on exciton-polariton transport for modes in the vicinity of the exciton energy. This model is applicable to any solid possessing dipole-active excitons.

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It has recently been demonstrated[1] that the characteristic doublet structure of the exciton polariton observed in the low-temperature photoluminescence spectrum of high-quality, nominally undoped GaAs grown by a variety of techniques is not of an intrinsic nature as generally thought. Previously, this doublet and similar doublets in other direct-gap compound semiconductors have been attributed to luminescence from the upper and lower polariton branches,[2] reabsorption,[3] and exciton-electric scattering.[4] However, in Ref. 1 it was shown in a series of samples that, as the concentration of shallow donors decreased, the line shape of the exciton-polariton emission changed from a wide asymmetric doublet, to a narrower symmetric doublet, to a single asymmetric peak as illustrated in Fig. 1. Although it was clear that this dramatic change was correlated with the presence of residual donors and was thus of an extrinsic nature, the physical mechanism giving rise to this effect was not obvious. Other workers[5] have suggested, without calculation or direct experimental evidence, that elastic scattering of excitons by residual impurities is the dominant scattering mechanism of exciton polaritons in GaAs at low temperatures. We now present a model of exciton-polariton photoluminescence line shapes that is based on this mechanism, i.e., on the elastic scattering of excitons from neutral impurities, and on the polariton nature of the exciton. We obtain very good agreement with line shapes observed, not only in samples with varying amounts of donors, but also in samples whose photoluminescence spectra are dominated by acceptor-related peaks. These results are direct evidence for the importance of scattering processes in exciton transport in GaAs and provide additional confirmation for the polariton nature of the exciton. However, our model is general in that it applies not just to GaAs, but to any solid with dipole-active excitons.

Nominally undoped GaAs epitaxial layers were grown several microns thick by molecular-beam epitaxy (MBE).[6] Photoluminescence spectra at 5 K were obtained by standard techniques.[1] The excitation intensity (100 mW/cm^2) was kept low enough to prevent heating or saturation effects. Figure 1 illustrates the range of spectra obtained in these experiments. In all samples the photoluminescence was dominated by free-to-bound transitions involving car-

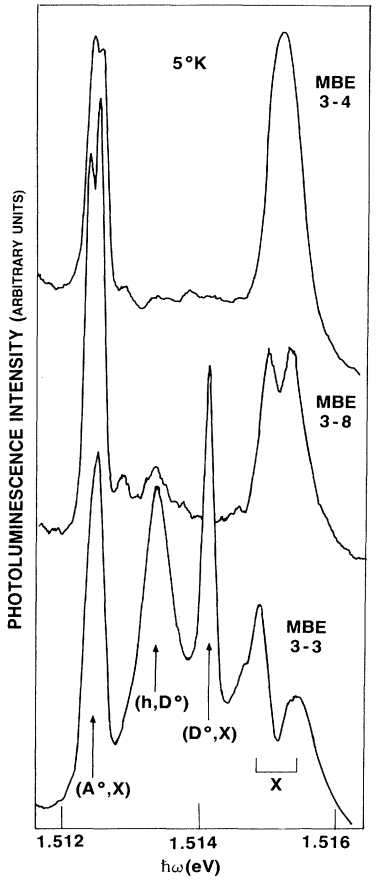

FIG. 1. 5-K photoluminescence spectra of three MBE-grown, nominally undoped, GaAs samples with varying concentrations of donors. The labeled structures are as follows: (A^0,X), excitons bound to neutral acceptors; (h,D^0), free-to-bound transitions involving neutral donors; (D^0,X), excitons bound to neutral donors; and X, exciton-polariton emission.

bon acceptors (at about 1.493 eV, not illustrated) which is consistent with Hall-effect measurements that indicate p-type material at room temperature with a net hole concentration in the range of 10^{15} cm^{-3}. Contact problems prevented temperature-dependent measurements. The donor concentration [as determined by the photoluminescence intensities of the donor-related peaks, (h,D^0) and (D^0,X), relative to the free-exciton structure[1]] varied widely in the three samples of Fig. 1: from a relatively large concentration in MBE 3-3, to moderate in MBE 3-8, to negligible in MBE 3-4. In step with these variations, the exciton-polariton line shape, labeled X, underwent the pronounced changes outlined above. Any postulated model must explain not only why the presence of donors leads to a dip, but also why no dip is observed if only acceptors are present, as in MBE 3-4.

Since the presence of shallow donors is necessary for the appearance of the doublet in the exciton-polariton photoluminescence line shape, a mechanism connecting shallow donors to exciton polaritons is required. Temperature-dependent photoluminescence studies indicated that the effect of exciton trapping on the line shape is negligible since the doublet was also present at higher temperatures at which impurity-bound excitons had been thermally ionized. Therefore, scattering is the operative mechanism. However, scattering by such massive atoms will be elastic at the small kinetic energies relevant to this problem. In addition, ionized impurities will be neutralized at low temperatures by the electrons and holes generated by the exciting laser beam and thus only neutral-impurity scattering need be considered. Therefore, our model is based on the elastic scattering of exciton polaritons by neutral donors and, in addition, neutral acceptors which cannot be neglected *a priori*.

Let us consider how the intrinsic line shape, deep in the bulk, is modified as the exciton polaritons progress toward the surface of the crystal and external detection. When the GaAs crystal is excited by above–band-gap energy radiation, electron-hole pairs diffuse into the crystal and lose energy through interactions with phonons, impurities, defects, etc. Once their energy is reduced below the band-gap energy, exciton polaritons, coupled exciton-photon modes, are the normal propagating modes in the crystal.[7] In GaAs, the exciton-polariton dispersion curve can be described in a two-branch model: Branch 1 begins at the longitudinal exciton energy, $\hbar\omega_L$, and quickly becomes photonlike at higher energies; branch 2 is photonlike below $\hbar\omega_L$ and excitonlike above $\hbar\omega_L$. Exciton polaritons continue to suffer energy loss through inelastic processes until their energies reach the vicinity of the transverse exciton energy, $\hbar\omega_T$, where they accumulate in the "bottleneck"[8] region as a consequence of competing loss mechanisms and are "quasithermalized." The "intrinsic" line shape deep in the bulk is proportional to the population distribution of these exciton polaritons in the bottleneck region. This distribution will be altered by inelastic and elastic scattering processes as these modes move through the crystal. Although elastic scattering processes cannot directly change the energy of the modes, they are important since they delay the arrival of the exciton polaritons at the surface. This effectively permits more time for inelastic processes, such as interactions with phonons, defects, etc., which do redistribute the population, to occur.[9] The elastic-scattering cross section of excitons from impurities is expected to depend on the kinetic energy of the exciton. The kinetic energy, in turn, will be a strong function of the exciton-polariton energy because of the rapid variation of mode group velocity near the exciton energy.[7] Thus the elastic-scattering cross section of exciton polaritons will vary significantly in the vicinity of the exciton energy, producing a strong variation in the total loss and thus a modification of the intrinsic line shape. The exact change in line shape will be a function of the dependence of the elastic-scattering cross section on the kinetic energy of the exciton polariton and need not be identical for donors and acceptors since these impurity atoms possess different scattering potentials.

We have calculated the elastic-scattering cross section for excitons from neutral donors and acceptors using partial-wave analysis. This is relevant to exciton polaritons since elastic scattering of exciton polaritons is dominated by the exciton component of the mode. The details of this calculation will be presented elsewhere.[10] The result of interest to this discussion is that the kinetic energy dependences of the cross sections for donors and acceptors are significantly different. This difference comes about in a fundamental way and can be intuitively understood. The magnitude of donor scattering is expected to be larger than that of acceptor scattering since the Bohr radius of a neutral donor is about an order of magnitude larger than that of an acceptor because of differences in the effective masses of electrons and holes. Thus a neutral donor is a larger "target." Further, the shape of the interaction potential between an exciton and a neutral impurity (that is, the magnitude and sign as a function of approach distance) is dependent on impurity type. In fact, for short distances, the interaction potential is repulsive for donors and attractive for acceptors.[10] Thus, it is not surprising that the shapes and magnitudes of the neutral-donor and -acceptor scattering cross sections as a function of exciton-polariton energy, given in Fig. 2, are quite different. Note that there is no direct physical reason why exciton polaritons on branch 2 near $\hbar\omega_L$ experience a maximum in cross section for scattering from donors while there is a local

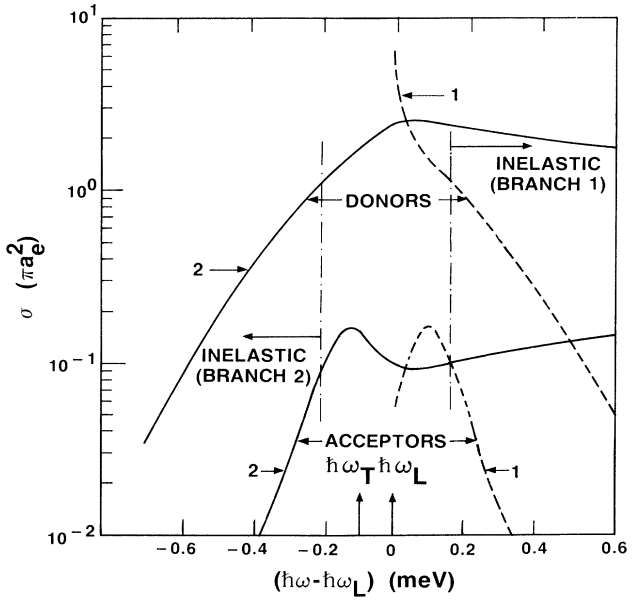

FIG. 2. Elastic-scattering cross section (in units of π times the square of the Bohr radius) of exciton polaritons in GaAs from neutral donors and acceptors as a function of the exciton-polariton energy relative to the longitudinal exciton energy, $\hbar\omega_L$, in millielectronvolts. The upper and lower polariton branches are denoted by 1 and 2, respectively. $\hbar\omega_T$ is the transverse exciton energy. The dash-dotted vertical lines delimit the energy range for each branch for which only elastic scattering is important.

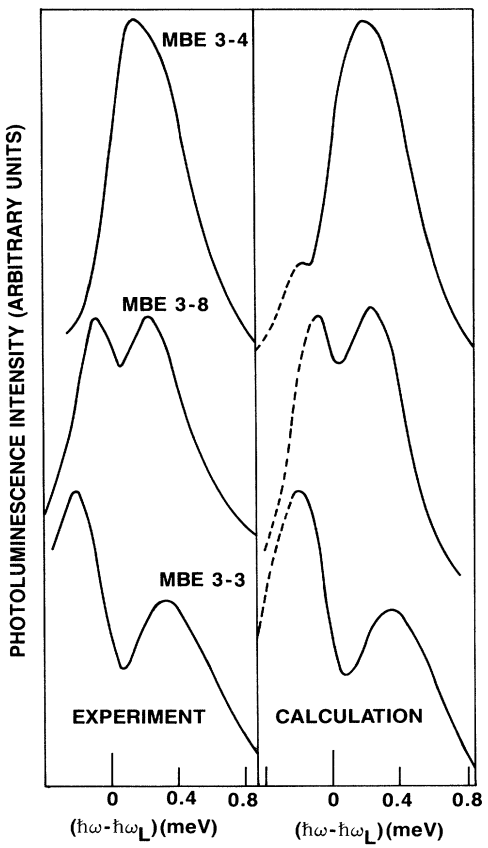

FIG. 3. Experimental and calculated exciton-polariton photoluminescence spectra for the samples in Fig. 1. The values on the abscissae are given relative to the energy of the longitudinal exciton, which has been determined from resonant Brillouin-scattering experiments (Ref. 7).

minimum for scattering from acceptors. This difference is an indirect consequence of the model. However, a large effect on calculated photoluminescence line shapes is produced by this difference and this is observed experimentally. The curves in Fig. 2 were obtained by first determining the kinetic energy of the exciton polariton for each value of energy using $E_q = 0.5MV_g^2$, where M is the exciton effective mass and V_g is the exciton-polariton group velocity, and then calculating the elastic-scattering cross section for that E_q. The range of interest is to the right of the dash-dotted line for scattering of modes on branch 2 and to the left of the dash-dotted line for scattering of modes on branch 1. In other regions inelastic scattering from impurities is possible since the kinetic energy of the mode is sufficient to excite the neutral impurity. Within the range of applicability of this model, however, significant differences in the dependences of the elastic-scattering cross section on exciton-polariton energy are evident and these will give rise to differences in the effects of this scattering on the line shapes of the exciton-polariton photoluminescence emission. Exciton-polariton modes which possess large cross sections will be strongly redistributed. Thus a "peak" in the scattering cross section will give rise to a "dip" in the photoluminescence line shape.

In order to compare this model with the experimental spectra of Fig. 1, we have assumed, for simplicity of fitting, a Gaussian for the intrinsic line shape of exciton-polariton emission from the "bottleneck" region on the lower exciton-polariton branch (branch 2) deep in the bulk. We further assumed that this line shape includes effects due to the energy-dependent transmission coefficient at the sample surface. There is negligible emission from modes on branch 1 since, even with these relatively low impurity concentrations, there is significant "thermalization" and most of the population resides in the "bottleneck" region on branch 2. For sample MBE 3-3 whose exciton photoluminescence is dominated by donors (which have larger elastic scattering cross sections than acceptors), we assumed a broader Gaussian (FWHM = 0.9 meV) than for MBE 3-4 in which donor scattering and, thus, thermalization is much weaker (FWHM = 0.61 meV). Since we had no direct measure of absolute donor concentrations in these samples, we took this into account

indirectly by matching the depth of the "dip" in the calculation to the experimental "dip."

The results of the fitting are presented in Fig. 3 together with the experimental spectra. The dashed lines in the calculated spectra represent regions in which inelastic processes must be included. These would have the effect of reducing the calculated intensities, thus, for example, reducing or eliminating the low-energy shoulder on the calculated spectrum of MBE 3-4. For sample MBE 3-4 whose photoluminescence spectrum is dominated by acceptor peaks, the calculation, assuming only acceptor scattering, produces a narrow, single asymmetric peak. On the other hand, for sample MBE 3-3 whose photoluminescence is dominated by donor peaks, the calculation, assuming only donor scattering, produces a broad doublet, characteristic of most high-purity GaAs.[1] Sample MBE 3-8 is an intermediate case. There are enough acceptors present to produce a narrow asymmetric underlying line shape as in sample MBE 3-4. However, there are sufficient donors still present to generate the characteristic "dip." It should be pointed out that the precise position of the dip, within our model, depends only on the shape of the exciton-polariton dispersion curve and the cross section as a function of kinetic energy and is independent of the magnitude of the dip. A deeper dip implies a larger concentration of neutral donor scattering centers, and, consequently, a broader Gaussian whose peak is shifted to lower energies. Thus as the donor concentration increases, the exciton-polariton line shape changes from a symmetric (MBE 3-8) to an asymmetric doublet (MBE 3-3) while the dip position remains fixed.

The overall agreement between the experimental and the calculated spectra provides convincing evidence for the viability of the model and thus for the importance of elastic scattering from neutral impurities in determining the line shape of the exciton-polariton photoluminescence emission in GaAs. Furthermore, it is clear that this scattering mechanism dominates transport for exciton-polariton modes near the exciton energy at low temperatures. Similar processes are expected to take place in other solids which possess dipole-active excitons and may account for the observed line shapes in, for example, ZnTe.[11]

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[9]In this picture, line shape will be a function of the thickness of the sample, i.e., the distance that the exciton polariton must travel to the surface. This dependence has been observed: W. L. Bloss, E. S. Koteles, E. M. Brody, B. J. Sowell, J. P. Salerno, and J. V. Gormley, to be published.

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