Comment on "Optically detected magnetic resonance in oxygen-doped GaP"

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This Comment presents a brief summary of the experimental inconsistencies found in a recent report by Godlewski and Monemar on optically detected magnetic resonance of the oxygen defect in GaP.

Substitutional oxygen in GaP is a point defect which for a number of semiconductor theorists represents the Rosetta stone of the theory of deep impurities.[1] The wealth of experimental data collected over the past 20 years allowed theorists to test a number of refined theoretical models. Research into this specific defect accelerated after 1978 when Morgan[2] reinterpreted the ir spectra of Dean and Henry[3] and suggested that the radiative transitions involved a second electron bound to O in GaP. In 1979, Gal *et al.*[4] reported experimental data which indicated that an excited spin-triplet state is responsible for the well-known 0.84-eV emission line of this defect, thereby supporting Morgan's model. In 1981 another luminescence band was discovered[5] which was also definitely shown to involve O through an isotope shift in the 0.52-eV no-phonon line.[6] To explain the origin of the triplet spin signal, Gal, Cavenett, and Dean[5] proposed in 1981 that spin-dependent Auger recombination takes place in the two-electron state (O^-) of the defect. This model was able to describe all of the known experimental observations regarding the optically detected magnetic resonance (ODMR) and photoluminescence (PL) data.

Recently, Godlewski and Monemar (GM) (Ref. 7) reported a new interpretation of the ODMR spectra in GaP:O. Their experimental results do not contain new or additional information; however, they claim that the spin-triplet resonance is *not* related to oxygen but rather to a Ga interstitial complex. Since this proposition would have significant consequences for the theory of deep states, it is important to highlight the experimental inconsistencies of GM's arguments.

There are a number of experimental observations reported in the literature indicating the *active* involvement of oxygen in the resonance process. Some of these are the following.

(a) The ODMR triplet resonances show strong polarization dependence on the 0.841-eV emission (which was proven repeatedly to be O related[1]) while no polarization dependence was observed on any of the other emission bands.[4]

(b) The ODMR signal was shown by Dawei and Cavenett to have the *same* monoclinic symmetry as was observed for the oxygen-related 0.528-eV no-phonon line.[8]

(c) "Level crossing" was only observed on the oxygen-related 0.841-eV emission.[4]

(d) It should also be noted that *only* GaP samples containing oxygen show the reported ODMR resonances.

Each of the above observations strongly suggests the direct involvement of oxygen in the ODMR process; however, GM seem to have disregarded all these data and have not commented on any of these published results.

The only notable result produced by GM to suggest that a center other than oxygen is responsible for the triplet resonance is their observation that the spectral dependence of the triplet ODMR resonance does not coincide with the oxygen-related donor-acceptor photoluminescence band.[7] Dawei and Cavenett[8] have measured the spectral dependence of the ODMR as well as that of the PL emission and have shown that if the spectral dependence of the *nonresonant* background is subtracted from the resonant signal (in order to obtain the undistorted spectra) the two spectra *do* peak at the same photon energy. The nonresonant background (the origin of which is not clear) is sample dependent and has to be measured for every sample. The lack of this procedure by GM could explain their results. GM's argument about the higher resolution of their spectra[7] is difficult to follow, since they do not quantify the spectral resolution of their measurements. In fact, they use a smaller monochromator than used by Dawei and Cavenett.[8]

It is also important to emphasize that in the case of donor-acceptor transitions, the ODMR and the PL spectra do not necessarily have to coincide. It is well known that donor-acceptor transitions involve donors and acceptors with varying spatial separation, resulting in pairs with diverse lifetimes and exchange interactions. Since ODMR is a sensitive function of lifetimes and/or exchange interactions, the spectral distribution of the ODMR signal can become quite different from that of the PL band. This was clearly demonstrated by Cox *et al.*[9]

We believe that until these issues are thoroughly discussed, GM's contribution will only confuse matters.

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