

to the relatively large concentration of impurities and changes in the force constants near them.

The resonant mode occurs at approximately one-half of the maximum phonon energy of 42 meV in Cr. From Eq. (2) and the phonon dispersion curves<sup>5</sup> it should be possible to calculate the frequency shifts to compare with experiment. We have observed evidence for resonant modes at approximately the same frequency in other branches of the phonon spectrum, and we are extending our preliminary experiments to make a detailed study of these.

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## OPTICAL ABSORPTION BY EXCITONS IN A STRONG ELECTRIC FIELD

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In this Letter we describe a nonperturbative treatment of a hydrogenic two-body system in the presence of a strong electric field. Our major results are (1) the demonstration that asymptotically for large negative energies the presence of the Coulomb interaction does not materially alter the optical absorption in an electric field alone, and (2) the calculation of the widths and energies of the low-energy exciton peaks as a function of the electric field strength. A number of speculations concerning the above results have appeared in the literature.<sup>1,2</sup> We present their first quantitative calculation. Such a calculation is needed to distinguish structure due to excitons from that due to critical points in electric-field-modulated optical reflectance<sup>2,3</sup> and transmission<sup>4</sup> data. In the course of our analysis, we perform the first exact solution of Schrödinger's equation for a potential, other than a square barrier, which exhibits bound states in the continuum.

If the interaction between the optical field and the solid is treated in first-order perturbation theory, then in the absence of other external fields it has been shown by Elliot<sup>5</sup> that the calculation of the optical-absorption tran-

sition probability reduces to evaluating the matrix element

$$M = \sum_{\vec{k}_e, \vec{k}_h} \Psi_{\vec{k}_e, \vec{k}_h, j, j'}^{\vec{K}, n} I_{jj'}(\vec{k}_e, \vec{k}_h, \vec{\xi}, \vec{q}), \quad (1a)$$

$$I_{jj'}(\vec{k}_e, \vec{k}_h, \vec{\xi}, \vec{q})$$

$$= \int d^3r \psi_{\vec{k}_e, j}^+(\vec{r}) e^{i\vec{q} \cdot \vec{r}} \left( \frac{i e \hbar}{m} \right)_{\vec{\xi}} \cdot \vec{\nabla}_{\vec{k}_h, j'}(\vec{r}), \quad (1b)$$

in which  $\vec{q}$  is the wave vector and  $\vec{\xi}$  the polarization vector of the light wave;  $\vec{k}_e$  and  $\vec{k}_h$  are the wave vectors of the electron and hole from bands  $j$  and  $j'$ , respectively; the  $\psi_{\vec{k}, j}$  are their one-electron wave functions, and  $\Psi$  is the Fourier transform of an exciton wave function. The imposition of an external electric field alters both the expansion coefficients,  $\Psi$ , and the one-electron wave functions,  $\psi$ . The effects of the electric field on  $\psi_{\vec{k}, j}$ , as reflected in the integral (1b), are thought to be small.<sup>6</sup> As the integral is customarily treated as a variable parameter used to fit the experimental data,<sup>7</sup> we do not discuss it further.

For direct, allowed transitions between two

parabolic bands,<sup>5</sup> we must evaluate  $\varphi_E(0)$ , with  $\varphi_E(\vec{r})$  satisfying

$$\left[\frac{1}{2}\Delta - \mathcal{E}z + r^{-1} + E\right]\varphi_E(\vec{r}) = 0, \quad (2a)$$

$$\begin{aligned} \mathcal{E} &= |e|Fa_B^3\mu/\hbar^2 \\ &= 1.94 \times 10^{-10}(m_e/\mu)^2\epsilon_0^3 F(\text{V/cm}), \end{aligned} \quad (2b)$$

$$E = (\hbar\omega - E_g)/(\hbar^2/\mu a_B^2), \quad (2c)$$

in which  $F$  is the external field strength,  $a_B$  is the Bohr radius,  $\epsilon_0$  is the static dielectric constant,  $\mu$  is the hydrogenic (electron-hole) reduced mass, and  $E_g$  is the zero-field energy gap. It is well known that Eq. (2a) is separable in parabolic coordinates [ $\xi = r+z$ ,  $\eta = r-z$ ,  $\varphi = \tan^{-1}(y/z)$ ], with the result<sup>6</sup> for  $E < 0$

$$\varphi_E(\vec{r}) = f_1(\xi)f_2(\eta)e^{im\varphi}/(\pi/2)^{1/2}, \quad (3)$$

$$\begin{aligned} \frac{d^2 f_i}{d\rho_i^2} + \frac{1}{\rho_i} \frac{df_i}{d\rho_i} + \left[ -\frac{1}{4} - \frac{m^2}{4\rho_i^2} + \frac{n_i + \frac{1}{2}(1+|m|)}{\rho_i} \right. \\ \left. + \frac{(-1)^i n^3 \mathcal{E} \rho_i}{4} \right] f_i = 0, \end{aligned} \quad (4a)$$

$$n = (-2E)^{-1/2}, \quad (4b)$$

$$n_1 + n_2 + 1 = n - |m|, \quad (4c)$$

$$\rho_1 = \xi/n; \rho_2 = \eta/n. \quad (4d)$$

From the indicial equation we find that for the wave function not to vanish at the origin, we need to consider only  $m=0$ . Although Eqs. (4) cannot be reduced to a two-term recursion relation, they were solved long ago in the weak-field limit ( $\mathcal{E} < 10^{-6}$ ) by a variant of perturbation theory and the results applied to the description of Stark shifts in atomic hydrogen.<sup>8</sup> However, in a discussion of electric field effects on exciton levels in semiconductors (e.g. GaAs), one must recognize that even modest fields ( $F=10$  V/cm) correspond to values of  $\mathcal{E}$  ( $\mathcal{E}=9 \times 10^{-4}$ ) far in excess of those ( $\mathcal{E} < 10^{-6}$ ) associated with third-order Stark shifts in hydrogen. Therefore, such a discussion requires an approach which a priori incorporates the influence of autoionization on even the lowest energy eigenfunctions (4a). We propose as a suitable starting point a model potential consisting of the Coulomb potential near the origin and the electric field far from the origin.

We obtain this potential by introducing a cut-off distance,  $x_0$ , equal to that at which the separate contributions to the  $\rho_2$  effective potential<sup>8</sup> are equal (when  $n_1 = n_2 = 0$ ):

$$\rho_1^{(0)} = \rho_2^{(0)} = x_0/n, \quad (5a)$$

$$x_0^3 - 2\mathcal{E}^{-1}x_0 - \mathcal{E}^{-1} = 0. \quad (5b)$$

The corrections to Stark shifts obtained from the model are small if  $n^3\mathcal{E} \gg 1$ . Thus, it is applicable in the interval  $0 > E > -\mathcal{E}^{2/3}$ . In particular, it describes the entire energy spectrum when  $\mathcal{E} \geq 1$  and the large- $n$  quasistationary states for any value of  $\mathcal{E}$  (the WKB treatment<sup>8</sup> of these states is rendered inapplicable by the boundary conditions). We next derive the absorption boundary-value problem and present some results obtained by use of our strong-field model potential.

In the presence of an electric field, Eqs. (4) have a continuous energy spectrum of both positive- and negative-energy eigenvalues and a discrete spectrum of eigenvalues for  $n_1$ . The eigenvalues of  $n_1$  are obtained from the solution to a transcendental equation and are labeled by the integral index  $i > 0$  such that  $n_1^{(i)} > n_1^{(j)}$  is denoted by  $i > j$ . The boundary conditions on the  $n_1$  quantum numbers for the optical-absorption problem may be defined by considering the zero-field limit in which both  $n_1$  and  $n_2$  are non-negative integers.<sup>8</sup> The parabolic-coordinate solution for the Coulomb field alone is well known<sup>8</sup> and can be related to the spherical-coordinate solution via the addition theorem for Laguerre polynomials.<sup>9</sup> If  $R_{nlm}(\vec{r})$  is the normalized solution in spherical coordinates and  $\psi_{n_1, n_2, n, m}(\vec{r})$  is the normalized solution in parabolic coordinates, we find

$$\begin{aligned} |R_{n00}(0)|^2 &= \frac{1}{n} \left| \sum_{s=0}^{n-1} \psi_{s, n-s-1, 0}(0) \right|^2 \\ &= \sum_{s=0}^{n-1} |\psi_{s, n-s-1, 0}(0)|^2. \end{aligned} \quad (6)$$

Thus we interpret a single  $l=0, m=0$  exciton line of intensity  $\pi^{-1}n^{-3}$  in spherical coordinates as the confluence of  $n$  lines each of intensity  $\pi^{-1}n^{-4}$  in parabolic coordinates. When the electric field is applied, the lines not only split up into  $n$  components (Stark effect), but also the energy spectrum becomes continuous. Each value of  $n_1^{(i)}$  gives rise to a "series" of quasistationary levels, one level being associated

with each zero-field level for  $n_0 > i$ . (We now denote the zero-field quantum number  $n$  by  $n_0$  because in the presence of the field  $n$  becomes a continuous variable  $n \geq 0$ .) Furthermore, each value of  $n_1^{(i)}$  contributes to the absorption for an arbitrary value of the energy ( $n$ ).

Writing the  $m = 0$  eigenfunctions (3) as

$$\varphi_{n_1, E}(\vec{r}) = (\xi n)^{-1/2} \chi_{n_1, E}(\xi) \chi_E(\eta), \quad (7a)$$

we find that the strength function for the optical-absorption problem becomes

$$|\varphi_E(0)|^2 = \sum_{i=0}^{\infty} |\varphi_{n_1^{(i)}, E}(0)|^2, \quad (7b)$$

$$\int_0^{\infty} \xi^{-1} |\chi_{n_1, E}(\xi)|^2 d\xi = 1, \quad (7c)$$

$$\int_0^{\infty} \chi_E(\eta) \chi_E(\eta) d\eta = \delta(E' - E), \quad (7d)$$

in which we have neglected nondivergent terms in the normalization integral. For weak fields, near the energy of one of the quasistationary levels, one term in (7b) is much larger than the sum of the remaining terms, and we find an absorption peak analogous to the zero-field exciton lines.

The details of computing  $\varphi$  will be published later.<sup>10</sup> Here we merely note that in the limit  $n \rightarrow 0$ , we can use integral values for  $n_1^{(i)}$  and explicitly evaluate the sum in (7b) in terms of asymptotic forms of Kummer<sup>11</sup> functions and Airy functions<sup>12</sup> to obtain

$$|\varphi_E(0)|^2 \xrightarrow{n \rightarrow 0} \pi A^2 \exp \left\{ -\frac{2}{3n^3 \mathcal{E}} [1 + c_1 n + c_2 n^2 + \dots] + c_0 \ln(n) \right\}, \quad (8a)$$

$$\pi A^2 = (2\mu)^{2/3} / (|e|F)^{1/3} \hbar^{4/3}. \quad (8b)$$

The  $c_1 n$  terms result from the dense spectrum of large  $n_1^{(i)}$  values, and  $c_2 n^2$  terms from the isolated low-lying  $n_1^{(i)}$  values. All of the  $n_1^{(i)}$  values contribute to the  $c_0 \ln(n)$  term. The important aspect of (8) is that the leading term in the brackets is unity so that the Coulomb interaction causes no asymptotic shift of the Franz-Keldysh band edge. This conclusion is independent of choice of  $x_0$  [provided  $x_0 < (n^2 \mathcal{E})^{-1}$ ], although the values of the  $c_i$  depend on  $x_0$ .

Near the zero-field band gap, the electric field causes two major modifications in the absorption spectrum. First, it splits (for  $n_0 > 1$ ), shifts, and broadens the exciton peaks and eventually makes them disappear. Second, it lowers the energy at which an electron-hole

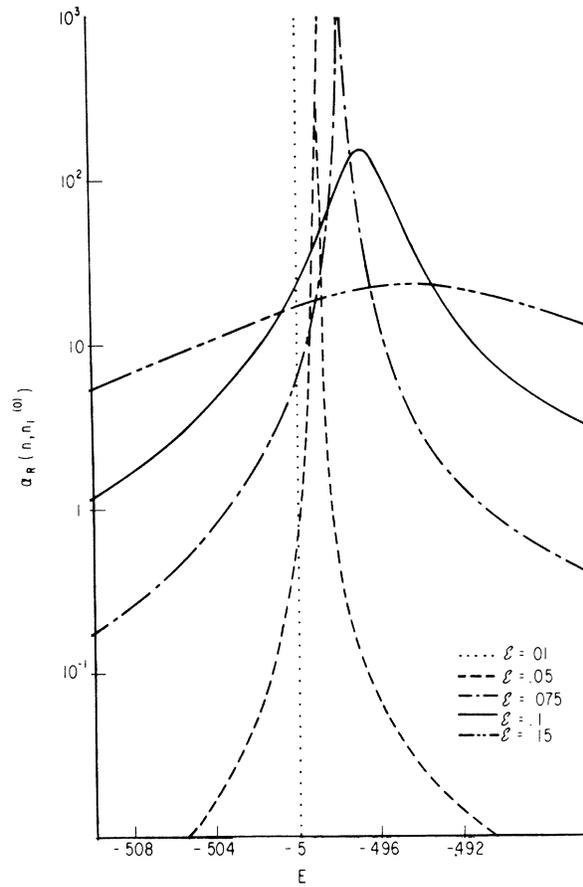


FIG. 1.  $\alpha_R(n, n_1^{(0)})$  as a function of the dimensionless energy parameter  $E$ . Energy is measured in units of  $E_0 = \hbar^2 / \mu a_B^2$ . For GaAs,  $E_0 = 11.3$  meV using the parameters of reference 7. Contributions to  $\alpha_R$  from  $\alpha_R(n, n_1^{(i)})$ ,  $i > 0$ , are not discernible on the scale used in the figure.

pair no longer needs to tunnel through a potential barrier in order to reach  $\vec{r} = 0$ . This latter result causes the absorption to reach its “continuum” edge at  $\hbar\omega < E_g$  in the presence of the electric field.<sup>13</sup> We calculate a dimensionless quantity  $\alpha_R(n, n_1) = |\varphi|^2 \mathcal{E}^{-1/3}$  which is proportional to the appropriate contribution to the absorption coefficient.<sup>14</sup> Figure 1 illustrates the line shape near the lowest energy exciton peak. The shift of the peak to higher energies is a consequence of the interaction of the oscillatory states outside the barrier with the quasistationary state inside the barrier. It is reduced (and for small  $\mathcal{E}$  reversed) by corrections to the model. The lowering of the “continuum” edge is reflected in the rise in the wings of the peak in  $\alpha_R$  as  $\mathcal{E}$  increases.

We conclude that although the electric field

does not alter the asymptotic edge,<sup>15</sup> there is a shift to lower photon energies of the apparent continuum absorption coupled with the disappearance of all exciton peaks at  $\mathcal{E} > \frac{1}{4}$ . Published GaAs absorption data<sup>16</sup> have been taken at temperatures which are too high to observe these effects. Preliminary data at liquid-nitrogen temperature<sup>17</sup> confirm the prediction of the shift in the continuum absorption in GaAs. A description of the experiments<sup>18</sup> in Cu<sub>2</sub>O requires the selection of  $m = 1$  in Eqs. (4). Our numerical calculations have not yet been extended to include this case.

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## INTERNAL FIELD OF Fe<sup>57</sup> IN Ni IN THE REGION OF THE CURIE POINT\*

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Recent calculations of the spontaneous magnetization and susceptibility of theoretical ferromagnets emphasize the importance of the region around the Curie point in analyzing specific models. The magnetization near  $T_C$  is generally described by the equation

$$M = \text{const} \times (1 - T/T_C)^\beta, \quad (1)$$

and a similar equation with exponent  $\gamma$  is used for the susceptibility. Numerical approximations of infinite series expansions yield  $\beta \approx 0.31$ <sup>1,2</sup> and  $\gamma \approx -5/4$ <sup>1,3</sup> for three-dimensional Ising ferromagnets, and  $\gamma \approx -4/3$ <sup>3-5</sup> for the Heisenberg model. These results, which are

relatively insensitive to the details of lattice structure, spin, and range of interaction, differ from the molecular-field model values  $\beta = \frac{1}{2}$  and  $\gamma = -1$ .

In measurements of the insulating ferromagnet EuS, Heller and Benedek<sup>6</sup> found that the zero-field resonant frequency of the Eu nuclei followed a  $\frac{1}{3}$ -power law ( $0.33 \pm 0.015$ ) over the range  $0.9 < T/T_C < 0.99$ . Experimental values for  $\gamma$  of metals appear to favor the Heisenberg model: for Fe,  $\gamma = -1.37 \pm 0.04$ <sup>7</sup> and  $-1.33$ <sup>8</sup>; for Ni,  $\gamma = -1.35 \pm 0.02$ <sup>9</sup> and  $-1.29 \pm 0.03$ <sup>10</sup>; and for Co,  $\gamma = -1.21 \pm 0.04$ .<sup>11</sup> Direct measurements of the spontaneous magnetization of metallic ferromagnets in the critical region have not