Interference resonance in two-phonon processes of de-excitation of the acceptors in tellurium

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(Submitted 30 October 1986)
Pis'ma Zh. Eksp. Teor. Fiz. **44**, No. 11, 532–534 (10 December 1986)

If the energy spectrum of a shallow impurity in the semiconductor has a large energy gap in which there is an additional level, the two-phonon de-excitation will exhibit a resonance when this level reaches the midpoint of the energy gap. This behavior probably accounts for the magnetic-impurity resonance in tellurium observed by Von Klitzing[3] [Solid State Electr. **21**, 223 (1978)].

The first excited state $|1\rangle$ of a shallow hydrogen-like impurity in a semiconductor is separated from the ground state $|0\rangle$ by a large energy gap E_{10} and is therefore metastable. Because the energy E_{10} is large, the phonon emitted in the transition $|1\rangle \to |0\rangle$ has a wavelength, $2\pi/q$, much shorter than the radius of the center, a, so that the matrix element of this transition is small in the parameter $qa \gg 1$. This metastability accounts for the fact that in strong electric fields $\mathscr{E}$, when the current flow is accompanied by an impact ionization of the impurities and recombination processes, an appreciable number of the impurities is in the excited state $|1\rangle$. The external magnetic field H changes the impurity spectrum and the band-electron spectrum. If the distance between the Landau levels in the band is the same as the energy gap E_{10}, the number of excited impurities will sharply decrease due to inelastic scattering of the carriers involving a transfer of energy E_{10} to them from the impurity. This resonance is responsible for the magnetic-impurity oscillations.[1,2]

Von Klitzing[3] detected an unusual "resonance" in the H dependence of a nonohmic resistance R (at $T = 2$ K, $\mathscr{E} \simeq 1$ V/cm). In tellurium the spectrum of a shallow impurity (bismuth, antimony, arsenic, phosphorus) is not hydrogen-like in the sense that instead of a single excited state, there are two excited states, $|1\rangle$ and $|2\rangle$, which are separated from the ground state $|0\rangle$ by a relatively large energy gap ($E_{20} \approx 1.1$ meV, $E_{10} \approx 0.4$ meV at $H = 0$). Special features of the magnetoresistance $R(H)$ are always observed[4] in such a field H when $E_{10} = E_{21} \equiv E_{20} - E_{10}$, i.e., when the level $|1\rangle$ lies halfway between $|2\rangle$ and $|0\rangle$. For example, when bismuth is the impurity and the field **H** is directed perpendicular to the crystallographic c axis, the resonance occurs in a field $H \approx 3.4$ T. A unique feature of this resonance condition is that it includes only the energy gaps from the impurity spectrum and that there are no energies other than those associated with the impurity. We will show below that at $E_{10} = E_{21}$ there occurs a resonant increase of the probability for the transition $|2\rangle \to |0\rangle$ with the emission of two phonons, which may account for the special features in the behavior of $R(H)$ observed by Von Klitzing.

The interaction of an impurity electron with the lattice vibrations is given by

 0021-3640/86/230687-03$01.00

$V = V_1 + V_2$, where V_1 is a one-phonon interaction, and V_2 is a two-phonon interaction. The transition $|2\rangle \to |0\rangle$ with the emission of two phonons, $\mathbf{q}$ and $\mathbf{q}'$, occurs in the second-order perturbation theory in V_1 and in the first-order perturbation theory in V_2. Accordingly, the matrix element of the transition can be written

$$M_{\mathbf{qq}'} = M^{(1)}_{\mathbf{qq}'} + M^{(2)}_{\mathbf{qq}'}, \tag{1}$$

where

$$M^{(1)}_{\mathbf{qq}'} = \langle 0; \mathbf{q}, \mathbf{q}' | V_2 | 2 \rangle, \tag{2}$$

$$M^{(2)}_{\mathbf{qq}'} = \frac{\langle 0; \mathbf{q}, \mathbf{q}' | V_1 | 1; \mathbf{q} \rangle \langle 1; \mathbf{q} | V_1 | 2 \rangle}{E_{21} - \hbar sq + i\hbar/2\tau_1} + \ldots . \tag{3}$$

Here the dots denote the term derived from the existing term through permutation of $\mathbf{q}$ and $\mathbf{q}'$, $\hbar sq$ is the phonon energy (s is the velocity of sound), and τ_1 is the lifetime of the state $|1\rangle$; $|\alpha;\ \mathbf{q},\ \mathbf{q}'...\rangle$ is the state of the system "impurity + lattice" when the phonons $\mathbf{q}$, $\mathbf{q}'$,... are in the excited state. The probability for the transition of interest to us is

$$w_{\mathbf{qq}'} = \frac{2\pi}{\hbar} |M_{\mathbf{qq}'}|^2 \, \delta(E_{20} - \hbar sq - \hbar sq'). \tag{4}$$

Since the matrix element $M^{(1)}$ contains the integral

$$\int d\mathbf{r} \psi_2^* (\mathbf{r}) \psi_0(\mathbf{r}) e^{i(\mathbf{q} + \mathbf{q}')\mathbf{r}}, \tag{5}$$

it is significantly greater than zero when $|\mathbf{q} + \mathbf{q}'| a \lesssim 1$, i.e., when

$$|\mathbf{q}| \approx |\mathbf{q}'| \approx E_{20}/2\hbar s. \tag{6}$$

The polar nature of the two-phonon interaction may be another reason that the matrix element $M^{(1)}$ is large when $|\mathbf{q} + '\mathbf{q}|$ are small.[5]

The matrix element $M^{(2)}$ is large for such phonons when the denominator in (3) is small, i.e., when

$$\hbar sq \approx E_{21}, \quad \hbar sq' \approx E_{10}. \tag{7}$$

or when the two-phonon transition $|2\rangle \to |0\rangle$ through a virtual intermediate state "reduces" to a series of two one-phonon transitions through an actual intermediate state. The square $|M_{\mathbf{q}-\mathbf{q}'}|^2$ appearing in the transition probability contains the interference term $M^{(1)}_{\mathbf{qq}'} M^{(2)*}_{\mathbf{qq}'}$ + c.c., which is large only if both conditions (6) and (7) hold, i.e., when $E_{21} \approx E_{10} \approx E_{20}/2$. This term is responsible, in our view, for the particular features of $R(H)$ observed in Fig. 3.

Relation (6) is valid within $\Delta q \sim 1/a$ and relation (7) is valid within $\Delta q \sim 1/\tau_1 s$. Consequently, the resonance width on the energy scale, i.e., the permissible deviation of the position of the level $|1\rangle$ from the midpoint of the gap E_{20} is

$$\Delta E \approx \hbar s/a + \hbar/\tau_1. \tag{8}$$

The lifetime τ_1 of the metastable state $|1\rangle$ is determined by the one-phonon processes. All available calculations of τ_1 are, however, for different type of centers (a hydrogen-like center,[6] acceptor in a cubic semiconductor,[7] donor in a multivalley semiconductor[8]). The time τ_1 can be estimated only crudely by analogy with other semiconductors where it is on the order of 10^{-7}–10^{-8} s. The first term in (8) is therefore the principal term. Substituting in it $s = 2\times10^5$ cm/s and $a = 150$ Å, we find $\Delta E \approx 0.1$ meV. Making use of the plots for the H dependence of the energy gaps E_{10}, E_{20}, and E_{21} (Fig. 9 in Ref. 4), we find from this value of ΔE that the resonance line width in the field should be about 0.7 T, which is in order-of-magnitude agreement with the resonance line width of the experimental curves (see e.g., Fig. 6 in Ref. 3).

The interference term in $|M_{\mathbf{qq}'}|^2$ cannot be larger than the sum $|M^{(1)}_{\mathbf{qq}'}|^2 + |M^{(2)}_{\mathbf{qq}'}|^2$. Since $M^{(1)}_{\mathbf{qq}'}$ and $M^{(2)}_{\mathbf{qq}'}$ are nearly independent of the field, the probability for the two-phonon transitions $w_{\mathbf{qq}'}$ at resonance can increase by no more than a factor of two. Since there are also other relaxation channels, it is not surprising that the resistance of tellurium at resonance varies very slightly (Fig. 1 in Ref. 3). The use of a modulation technique, however, makes it possible to detect these resonances with a large sensitivity margin.

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Translated by S. J. Amoretty