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Effect of charge-density waves on the magnetic susceptibility of niobium triselenide

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The magnetic susceptibility of a model charge-density wave (CDW) system is discussed. It is found that the orbital part of the susceptibility can be large and paramagnetic due to mixing of states above and below the CDW energy gaps by the magnetic field. The paramagnetic contribution is anisotropic and depends on the square of the sine of the angle between the magnetic field $\vec{H}$ and the CDW wave vector $\vec{Q}$. The relevance of these findings to recent measurements of the susceptibility of niobium triselenide is discussed.

I. INTRODUCTION

The novel properties of niobium triselenide ($NbSe_3$) have been the subject of considerable investigation in recent years. To date $NbSe_3$ is unique among the materials known to exhibit charge-density-wave (CDW) distortions in that it contains two, apparently unrelated CDW's, with transition temperatures marking their appearance at $T_1 = 142$ K and $T_2 = 58$ K. The periodicities of the two charge-density waves, [$\vec{Q}_1 = (0, 0.243, 0)$ and $\vec{Q}_2 = (0.5, 0.263, 0.5)$], have been directly observed by electron[1,2] and x-ray[3] diffraction. Of considerable interest have been the temperature[4] and (especially) the electric-field[5–7] dependences of the resistivity of $NbSe_3$. Electric-field-dependent effects in the thermopower[8] and the presence of periodic voltage fluctuations in the noise spectrum[9] accompanying the nonohmic resistivity have recently been discovered.

Interpretation of many experiments is difficult because of the lack of complete band structure information. Owing to the complicated crystal structure,[2,10] which has six formula units in the monoclinic unit cell, existing band structure calculations[11] do not allow determination of the Fermi surface, even in the undistorted state. Magneto-oscillatory transport measurements[12–14] are made at temperatures well below the CDW transitions, and therefore only give information about the Fermi surface for electrons in the distorted lattice. Moreover, without a calculated Fermi surface, interpretation of the measured Fermiology data is unclear.

One quantity of interest is the fraction of Fermi surface destroyed by the presence of the CDW energy gaps. From analyses of the resistivity Ong[15] and Ong and Monceau[7] concluded that the fractions of the Fermi surface affected by the CDW's were 20% at T_1 and 60% at T_2. The very small low-temperature electronic specific heat[16] (< 5 erg/g K) supports the conclusion that the remaining density of states at the Fermi surface is small when both CDW's are present. In order to elucidate further the density of states, Kulick and Scott[17] measured the static magnetic susceptibility of compressed powder samples of $NbSe_3$. While their analysis was complicated somewhat by an uncertainty in the core diamagnetism, they concluded (unambiguously) that if the changes in the susceptibility are interpreted in terms of the removal of density of states, then the fractions affected at the two transitions (and especially T_2) are significantly smaller from those found from the analysis of resistivity. They concluded that there must be an additional paramagnetic contribution to the susceptibility which accompanies the CDW's in order to resolve the discrepancy.

The purpose of this paper is to demonstrate that in the presence of a CDW which gaps a finite part of the Fermi surface there is an additional zero-temperature paramagnetic contribution to the susceptibility. (There are additional temperature-dependent effects which will be the subject of later study.) Moreover, this additional paramagnetism is dependent on the square of the sine on the angle between $\vec{H}$ and the CDW wave vector $\vec{Q}$. It is, thus, zero when $\vec{H}$ and $\vec{Q}$ are parallel. The anisotropy is a fortunate occurrence, because without band structure and Fermi surface information it is not possible to calculate the size of this contribution (except to demonstrate that it can be important). By performing measurements on aligned samples, it should be possible to measure this term and to properly determine the density-of-states effects.

II. MODEL FERMI SURFACE

The actual Fermi surface of $NbSe_3$ is not known and is likely to be very complicated with possibly several pockets of electrons and holes. In this paper we choose to simplify the calculation by confining

discussion to a model Fermi surface.

Consider a model system in which the electron energies in the absence of the CDW are given by

$$\epsilon_{\vec{k}} = \hbar^2[k_x^2/2m_x + (k_y^2 + k_z^2)/2m_y] \ . \tag{1}$$

For simplicity the effective masses for the $\hat{y}$ and $\hat{z}$ directions have been taken as equal. We take the wave functions in the absence of the CDW to be plane waves. The generalization to Bloch functions is not difficult, but is not essential to the discussion.

We now pretend that there is a CDW present so that each electron experiences a one-electron potential

$$V(\vec{r}) = G\cos\vec{Q}\cdot\vec{r} \ . \tag{2}$$

G is the CDW energy gap and $\vec{Q}$ is the CDW wave vector. For this discussion $\vec{Q}$ is chosen parallel to $\hat{x}$ so that m_x (m_y) is the electron mass characterizing dispersion parallel (transverse) to $\vec{Q}$. The essential feature of the energy in Eq. (1) is the finite dispersion transverse to $\vec{Q}$.

The CDW potential in Eq. (2) mixes the unperturbed state (with wave vector) $\vec{k}$ with $\vec{k} \pm \vec{Q}$ and produces gaps in the one-electron energy at $\vec{k} = \pm\frac{1}{2}\vec{Q}$. Using first-order perturbation theory, it is easy to find the wave functions and energies for states below and above the gaps. Because this procedure is well known, we do not catalog the results here but will call upon them when needed.

We wish to describe the case in which the Fermi surface contacts the CDW energy gaps in a finite area, and we take only states below the gaps to be occupied at zero temperature. With a change in variables,

$$\begin{aligned} &u = k_y/Q, \quad v = k_z/Q \ , \\ &w = (k_x + \tfrac{1}{2}Q)/Q, \quad k_x < 0 \ , \\ &w = (k_x - \tfrac{1}{2}Q)/Q, \quad k_x > 0 \ , \end{aligned} \tag{3}$$

the energy for an electron in a state below the gaps labeled by wave vector $\vec{k}$ is approximately

$$E_{\vec{k}} = (\tfrac{1}{2}\hbar^2Q^2)\{(u^2+v^2)/m_y + [w^2 + \tfrac{1}{4} - (w^2+\alpha^2)^{1/2}]/m_x\} \ , \tag{4}$$

where $\alpha \equiv m_x G/\hbar^2Q^2$. As in Fig. 1, let the radius of the (circular) neck, where the Fermi surface contacts the CDW energy gaps, be $\kappa_0 Q$. At the gap on the right, $k_x = \frac{1}{2}Q$ and

$$E_F = (\tfrac{1}{2}\hbar^2Q^2)[\kappa_0^2/m_y + (\tfrac{1}{4} - \alpha)/m_x] \ . \tag{5}$$

From Eqs. (4) and (5) the equation for the Fermi surface in terms of w and $\kappa \equiv (u^2+v^2)^{1/2}$ is

$$\kappa^2 = \kappa_0^2 + (m_y/m_x)[(w^2+\alpha^2)^{1/2} - \alpha - w^2] \ . \tag{6}$$

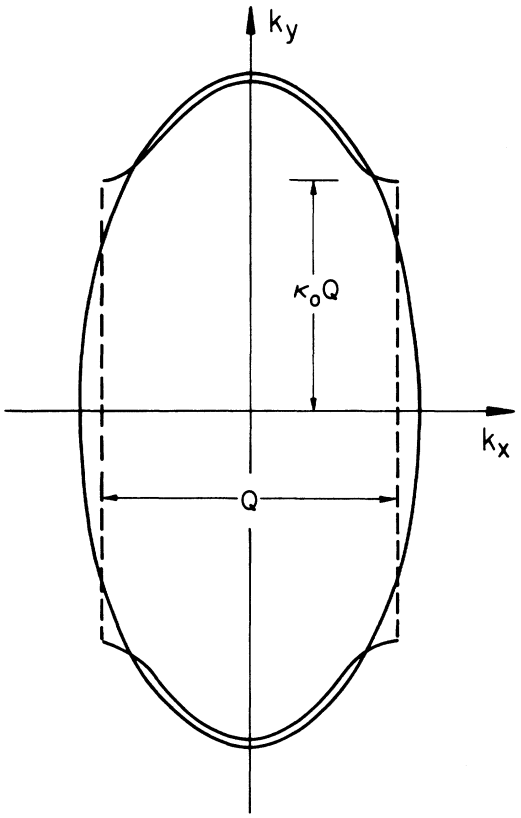

FIG. 1. Cross section of model Fermi surface. With no CDW present, the cross section is elliptical. With a CDW present, the Fermi surface contacts the CDW energy gaps in necks of radius $\kappa_0 Q$. Q is the CDW wave vector.

The number of electrons enclosed by the Fermi surface is easily found to be

$$\begin{aligned} N &= \left(\frac{Q^3}{2\pi^2}\right)\int^{1/2} dw\, \kappa^2 \\ &\cong \left(\frac{Q^3}{4\pi^2}\right)\left[\kappa_0^2 + \left(\frac{m_y}{m_x}\right)\left[\tfrac{1}{2}(\tfrac{1}{4}+\alpha^2)^{1/2} - \alpha^2\ln\alpha - \alpha - \tfrac{1}{12}\right]\right] \ . \end{aligned} \tag{7}$$

If the diameter of the unperturbed Fermi surface along the $\hat{x}$ direction is $2k_0$, then the number N_0 enclosed by the unperturbed Fermi surface is

$$N_0 = (k_0^3/3\pi^2)(m_y/m_x) \ . \tag{8}$$

It is easy to show for both the unperturbed and perturbed Fermi surfaces that

$$d^3k = (m_y/\hbar^2)\, dE_{\vec{k}}\, d\phi\, dk_x \ , \tag{9}$$

where ϕ is the usual angle in cylindrical coordinates.

III. SUSCEPTIBILITY

Before turning our attention to the main subject of discussion, for completeness we calculate the Pauli susceptibility of the model system. Using Eqs. (8) and (9) the Pauli susceptibilities of the unperturbed (χ_p^0) and perturbed (χ_p) Fermi surfaces are

$$\chi_p^0 = \mu_B^2 m_y k_0/\pi^2\hbar^2 \ , \tag{10}$$

$$\chi_p = \mu_B^2 m_y Q/2\pi^2\hbar^2 \ , \tag{11}$$

where μ_B is the Bohr magneton.

When the CDW is present, the Fermi surface is described by a number of parameters: N, E_F, κ_0, Q, G. (We assume k_0, m_x, and m_y are known for the unperturbed case.) Equations (5) and (7) give two relations among these. If the Fermi surface consists of only the one piece under discussion, then N must be equal to N_0. If there are additional pieces to the Fermi surface, then a repopulation of the various pieces may occur in going to the CDW state.

We now turn our attention to the orbital or diamagnetic susceptibility. The theory of the diamagnetic susceptibility of metals has been studied by many workers. It is convenient here to use the results of Misra and Roth[18] who considered the case of nearly free electrons and included the case in which electron states near the energy gaps are occupied. A convenient starting point is Eq. (6.6) of Misra and Roth modified for the present notation.[19] If coordinates are chosen such that the magnetic field is $\vec{H} = H\hat{z}\sin\theta + H\hat{x}\cos\theta$, $\vec{Q} = \vec{Q}\hat{x}$, and for the present $m_x = m_y$ is the free electron mass m, then the orbital susceptibility is

$$\chi_{\text{orb}} = \frac{\mu_B^2}{12\pi^3}\int d^3k\, f'(E_{\vec{k}}) + \frac{4\mu_B^2 m^2 Q^2 \sin^2\theta}{\pi^3\hbar^4}\left[\int d^3k\, f'(E_{\vec{k}})\frac{G^2}{[(Q^2+2k_xQ)^2+4\alpha^2]^{3/2}}\left(\frac{1}{6}-\frac{k_y^2}{[(Q^2+2k_xQ)^2+4\alpha^2]^{1/2}}\right)\right.$$
$$\left. + \frac{m}{\hbar^2}\int d^3k\, f(E_{\vec{k}})\frac{G^2}{[(Q^2+2k_xQ)^2+4\alpha^2]^2}\left(1+\frac{8k_y^2}{[(Q^2+2k_xQ)^2+4\alpha^2]^{1/2}}\right)\right] . \tag{12}$$

$f(E_{\vec{k}})$ is the Fermi occupation factor and $f' \equiv df(E)/dE$. A factor of 2 has been included to account for the two energy gaps.

It is convenient to label the first term on the right-hand side of Eq. (12) χ_1. This term is immediately evaluated as

$$\chi_1 = -\tfrac{1}{3}\chi_p \ , \tag{13}$$

where χ_p is the Pauli susceptibility. The case $m_x \neq m_y$ does not appear to have been considered in the literature and is not of primary interest here since it is reasonable to assume that Eq. (13) would be modified by some suitable average of the masses.

It is straightforward to examine the other (anisotropic) terms on the right-hand side of Eq. (12) by expanding the integrand in a power series in $2mG/\hbar^2Q^2 \cong G/4E_F$. In each term the integration leads to an additional factor of order $G/4E_F$ due to the small number of electrons in the strongly perturbed regions near the energy gaps (see Fig. 1). Consider, for example, the last term in Eq. (12). The leading power of G is obtained near the gap where $Q^2+2k_xQ \lesssim \alpha$. The integrand is of order G^{-3}, and the additional factor of G from the integration makes the leading term of order G^{-2}. It is easy to show that all the other terms in Eq. (12) contribute at most to order G^{-1}. We, therefore, confine our discussion to evaluating the last term in Eq. (11), since it will dominate for small G, and label it χ_2.

An alternative derivation of χ_2 is given in the Appendix. There it is shown that χ_2 arises from the change in energy due to mixing by the magnetic field of states (with the same wave-vector label) in different bands. Hence, χ_2 can be called the interband orbital susceptibility. The origin of the positive (paramagnetic) sign of χ_2 is easily seen by the following argument. When two states are mixed in perturbation theory, the state with the lower energy is pushed still lower in energy by $-\Delta$, ($\Delta > 0$), while the higher one is raised by Δ. If only the lower state is occupied, the susceptibility is given by $\chi = -\partial^2 \times (-\Delta)/\partial H^2$ and is positive. χ_2 is an example of Van Vleck paramagnetism involving extended states.

In the Appendix the form of χ_2 for $m_x \neq m_y$ is derived with the result

$$\chi_2 = \left(\frac{32\mu_B^2 m^2 m_x^3 G^2 Q^2 \sin^2\theta}{m_y^2\hbar^6\pi^3}\right) \times \int d^3k\, f(E_{\vec{k}})\, k_y^2 [(Q^2-2k_xQ)^2+4\alpha^2]^{-5/2} . \tag{14}$$

It is not difficult to evaluate χ_2 to leading order in G to obtain

$$\chi_2 \cong \frac{4\mu_B^2 m^2 \kappa_0^4 N E_F (m_x\kappa_0^2/m_y + \frac{1}{6})\sin^2\theta}{3m_x m_y G^2 (m_x\kappa_0^2/m_y + \frac{1}{4})} \ , \tag{15}$$

where the expressions for E_F and N in Eqs. (5) and

(7) have been used.

As explained above it is not possible to estimate convincingly the size of χ_2 because there is little information on the various parameters involved. However, the ratio of χ_2 to the Pauli susceptibility is proportional to $(E_F/G)^2$ so that it would not be surprising to find a large χ_2 for small CDW energy gaps. An interesting feature of χ_2 is its dependence on m_y, which characterizes the curvature of the electron energy in a direction perpendicular to $\vec{Q}$. Within the context of the model Fermi surface chosen here for calculational expediency, increasing m_y serves to make the system more nearly one dimensional. From Eq. (15) $\chi_2 \sim m_y^{-1}$ and, thus, decreases as the system approaches one dimensionality. As noted in the Appendix, χ_2 is identically zero for a true one-dimensional system.

An important property of χ_2 is its anisotropy. From Eq. (15) χ_2 is seen to be zero when $\vec{H}$ is parallel to $\vec{Q}$. Thus, a measurement of the angular dependence of the susceptibility of carefully aligned samples would serve to determine χ_2. The remaining susceptibility would then give the desired information about the density of states at the Fermi surface.

From the result in Eq. (15), it would appear that χ_2 will diverge as $G \rightarrow 0$; i.e., the susceptibility is infinite in the limit that the CDW vanishes. The source of this unphysical result is the tacit assumption that only states below the CDW energy gaps are occupied. To examine more closely the $G \rightarrow 0$ limit, consider the case in which the Fermi surface consists of only this one piece. Since the unperturbed model Fermi surface has been taken to be an ellipse as in Fig. 1, states above the gaps will become filled as $G \rightarrow 0$. Filling of states above the gaps will begin when G is sufficiently small so that the energy of states above the gaps is less than that of the states below the gaps (at finite transverse wave vector) which become populated in the presence of the CDW. This is an important point which requires further study. Unfortunately, the simple model chosen for this study (although adequate for the discussion of magnetic field effects) is not sufficient for questions related to the energy of the CDW since the CDW state is not energetically favorable in the first place.

Although the $G \rightarrow 0$ limit cannot be discussed in detail, it is possible to see what will happen. The states above the gaps which become filled as $G \rightarrow 0$ contribute to the susceptibility with opposite sign (as is usual in perturbation theory) and cancel the most divergent behavior of the interband susceptibility. It is a tedious (although not a difficult) exercise to start with the complete expression, Eq. (12), for the orbital susceptibility and to demonstrate that the anisotropic part of χ_{orb} does go to zero as $G \rightarrow 0$ in the case of an isotropic effective mass. The generalization of Eq. (12) to anisotropic effective mass, the detailed study of the intraband terms in the orbital susceptibility, and an investigation of finite temperature effects is an important area for further research.

IV. CONCLUSION

It has been shown that when a CDW truncates a finite part of the Fermi surface, there can be a large, paramagnetic contribution χ_2 to the susceptibility. χ_2 arises due to the virtual mixing of states below and above the CDW energy gaps by the magnetic field and is an example of Van Vleck paramagnetism for extended states.

An important property of this paramagnetic contribution to the susceptibility is its anisotropy. It was found that χ_2 varies with the square of the sine of the angle between the magnetic field $\vec{H}$ and the CDW wave vector $\vec{Q}$. Thus, measurements of the anisotropy of the susceptibility for aligned samples can determine χ_2.

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APPENDIX

The purpose of this Appendix is to demonstrate that the orbital contribution to the susceptibility labeled χ_2 above arises due to the change in energy resulting from the (virtual) mixing of states above and below the CDW energy gaps by the magnetic field. It is convenient to use the Bloch[20,21] (or crystal momentum) representation. This method has been previously used by the author in a microscopic derivation of the equation of motion of a CDW in a magnetic field,[22] and further details of its application can be found therein.

If a general wave function Ψ is

$$\Psi(\vec{r}) = \sum_n \int d^3k \, f_n(\vec{k}) e^{i\vec{k}\cdot\vec{r}} u_{n\vec{k}}(\vec{r}) \qquad \text{(A1)}$$

in the coordinate representation, then in the Bloch representation

$$\Psi = [f_0(\vec{k}) f_1(\vec{k}) f_2(\vec{k}) \cdots] \qquad \text{(A2)}$$

is an (infinite) column vector. In this representation the coordinate operator $\vec{r}$, for example, is[20]

$$\vec{r} = il\nabla_{\vec{k}} + X_{nn'}(\vec{k}) \ , \qquad \text{(A3)}$$

where I is an (infinite) unit matrix and

$$X_{nn'}(\vec{k}) \equiv i \int d^3k\, u_{n\vec{k}}(\vec{r}) \nabla_{\vec{k}} u_{n'\vec{k}}(\vec{r}) \quad . \tag{A4}$$

Note that the off-diagonal matrix $X_{nn'}$ connects states with the same (reduced) wave vector $\vec{k}$ in different bands n and n'.

The Hamiltonian in the absence of the magnetic field is

$$H_0 = p_x^2/2m_x + (p_y^2 + p_z^2)/2m_y + G\cos\vec{Q}\cdot\vec{r} \quad . \tag{A5}$$

As above, $\vec{Q}$ is chosen parallel to $\hat{x}$. In the presence of the magnetic field, the Hamiltonian is

$$H = \frac{[p_x + eA_x(\vec{r})/c]^2}{2m_x} + \frac{[p_y + 2eA_y(\vec{r})/c]^2}{2m_y} + \frac{[p_z + eA_z(\vec{r})/c]^2}{2m_y} + G\cos\vec{Q}\cdot\vec{r} - e\phi(\vec{r}) \quad . \tag{A6}$$

$\vec{A}(\vec{r})$ and $\phi(\vec{r})$ are the vector and scalar potentials which are properly chosen[23] in the (time-dependent) Jones and Zener gauge[24]

$$\vec{A} = \tfrac{1}{2}\vec{H}\times(\vec{r} - \vec{v}_G t) \quad , \qquad \phi = (1/2c)(\vec{H}\times\vec{v}_G)\cdot\vec{r} \quad . \tag{A7}$$

$\vec{v}_G \equiv \nabla_{\vec{k}} E(\vec{k})/\hbar$ is the group velocity of an electron with energy $E(\vec{k})$.

If $\vec{H} = H\cos\theta\hat{x} + H\sin\theta\hat{z}$, the Hamiltonian at time $t=0$ (Ref. 25) is $H = H_0 + H'$, and

$$H' = (e/2m_x c)H\sin\theta y p_x + (e/2m_y c)H(\sin\theta x p_y - \cos\theta z p_y) + (e/2m_y c)H\cos\theta y p_z - (e/2c)(\vec{H}\times\vec{v}_G)\cdot\vec{r} \quad . \tag{A8}$$

The A^2 term has been dropped since it does not contribute to the final result.

The wave functions in the absence of the magnetic field are those which diagonalize H_0. The CDW potential $G\cos\vec{Q}\cdot\vec{r}$ can (for our purpose) be separated into two parts, one which produces the gap at $\vec{k} = \frac{1}{2}\vec{Q}$ and another which produces the gap at $\vec{k} = -\frac{1}{2}\vec{Q}$. The former can be diagonalized on the basis of $\vec{k}$ and $\vec{k} - \vec{Q}$ with the result for wave functions below and above the energy gaps

$$\phi_{\vec{k}} = e^{i\vec{k}\cdot\vec{r}}\cos\xi_{\vec{k}} - e^{i(\vec{k}-\vec{Q})\cdot\vec{r}}\sin\xi_{\vec{k}} \equiv u_{\vec{k}} e^{i\vec{k}\cdot\vec{r}} \quad , \tag{A9}$$

$$\Psi_{\vec{k}} = e^{i\vec{k}\cdot\vec{r}}\sin\xi_{\vec{k}} + e^{i(\vec{k}-\vec{Q})\cdot\vec{r}}\cos\xi_{\vec{k}} \equiv v_{\vec{k}} e^{i\vec{k}\cdot\vec{r}} \quad .$$

The energies of the states above and below the gaps are

$$E_{\pm}(\vec{k}) = \tfrac{1}{2}(\epsilon_{\vec{k}} + \epsilon_{\vec{k}-\vec{Q}}) \pm \tfrac{1}{2}[(\epsilon_{\vec{k}} - \epsilon_{\vec{k}-\vec{Q}})^2 + G^2]^{1/2} \quad , \tag{A10}$$

where $\epsilon_{\vec{k}}$ is given in Eq. (1). The coefficients in Eqs. (A9) are determined by the relation

$$\sin 2\xi_{\vec{k}} = G/[E_+(\vec{k}) - E_-(\vec{k})] \equiv G/W(\vec{k}) \tag{A11}$$

and are independent of k_y and k_z.

In the Bloch representation the effect of H' is to perturb the basis functions $u_{n\vec{k}}(\vec{r})$.[20,24] Before considering H' of Eq. (A8), it is instructive to consider the effect of two typical terms, $H_1 = C_1 x$ and $H_2 = C_2 x p_y$, where C_1 and C_2 are constants of no interest other than to give overall units of energy.

The effect of H_1 on, say, the basis function $u_{\vec{k}}$ and $v_{\vec{k}}$ of Eqs. (A9) is to mix them according to Eq. (A3). The perturbed basis functions which diagonalize $H_0 + H_1$ to first order are easily obtained. The perturbed function $u_{\vec{k}}^{(1)}$ is[20,24]

$$u_{\vec{k}}^{(1)} = u_{\vec{k}} + v_{\vec{k}} \frac{\langle v_{\vec{k}}/iC_1\,\partial/\partial k_x / u_{\vec{k}}\rangle}{E_-(\vec{k}) - E_+(\vec{k})} \quad , \tag{A12}$$

and $v_{\vec{k}}^{(1)}$ is obtained *mutatis mutandis*. Equation (A12) resembles the usual perturbation theory result except that the $u_{n\vec{k}}$'s appear rather than the $\phi_{n\vec{k}}$'s and x is replaced by $i\,\partial/\partial k_x$.

Similarly, under the action of H_2,

$$u_{\vec{k}}^{(1)} = u_{\vec{k}} + \hbar C_2 v_{\vec{k}} \frac{\langle v_{\vec{k}}/(ik_y + \partial/\partial y)\,\partial/\partial k_x / u_{\vec{k}}\rangle}{E_-(\vec{k}) - E_+(\vec{k})} \quad . \tag{A13}$$

Since the coefficients $\cos\xi_{\vec{k}}$ and $\sin\xi_{\vec{k}}$ in Eqs. (A9) are independent of k_y and k_z, several of the terms in H' do not contribute. Calculating the change in energy $\Delta E(\vec{k})$ to second order in H for an electron in a state below the gap using the perturbed function $u_{\vec{k}}^{(1)}$ obtains

$$\Delta E(\vec{k}) = -e^2 H^2 \hbar^6 G^2 k_y^2 Q^2 / 4 m_x^2 m_y^2 W^5 \quad . \tag{A14}$$

The expression for χ_2 in Eq. (14) follows by summing $\Delta E(\vec{k})$ over occupied states, multiplying by 2 to account for both gaps, and using the well known relation between χ and the total change in energy ΔE; i.e., $\chi \equiv -(\partial^2 \Delta E/\partial H^2)$.

It is worthwhile to note that for a truly one-dimensional conductor, $\Delta E(\vec{k})$ is identically equal to zero because matrix elements such as those in Eqs. (A12) and (A13) are zero when there is no dispersion perpendicular to the CDW direction. Thus, in a one-dimensional conductor the paramagnetic χ_2 accompanying the CDW is equal to zero. This result is in agreement with experimental measurements of the susceptibility of tetrathiafulvalene-tetracyanoquinodimethane.[26]

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