FIG. 2. A double chain system of $CuCl_4^-$ complexes. The circles represent the $CuCl_4^-$ and the interchain exchange paths are represented by dashed lines.

be considered constant throughout the crystal, but D is a function of position. Figure 2 shows a double chain with an exchange interaction between the chains. This kind of $CuCl_4^-$ chain structure occurs in bis-piperidinium tetrachlorocuprate,[5] $(C_5H_{10}NH_2)_2CuCl_4$. To make the D vector more obvious in Fig. 2, we have exaggerated an inequivalence between the chains by rotating one about its axis relative to the other by 180°. D still changes direction as we go down each chain and the intrachain D will have a zero average over the complete chain. However, the interchain D has a nonzero average or there will be a constant part of the D vector over the double chain system perpendicular to the chain.

We have shown that D is usually a function of positions in a crystal and there is at least one structure that exhibits a partially constant D. Earlier theoretical results[1,2] were based on the continuum approximation which assumes only very small changes in the Hamiltonian between adjacent crystal sites. Clearly, the D vector in most physical systems will not satisfy this approximation so we must be careful relating theoretical results to a physical system with a Dzyaloshinsky-Moriya interaction.

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COMMENTS

Comment on: "A quantum analog to the classical quasiperiodic motion"

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In a recent article[1] Hose and Taylor suggest that the conventional local stability condition of the Wigner Brillouin (WB) perturbation series may serve as a means of identifying quantum mechanical levels corresponding to classical quasiperiodic motion. Their criterion for quasiperiodic nature is

$$\frac{\langle \Psi_p | \frac{d}{dE} H_{eff}(E) | \Psi_p \rangle}{\langle \Psi_p | \Psi_p \rangle}\Big|_{E=\bar{E}} < 1 \tag{1a}$$

or alternatively

$$\eta \equiv \frac{\langle \Psi_p | \Psi_p \rangle}{\langle \Psi | \Psi \rangle} > 0.5 . \tag{1b}$$

Here $\Psi_p = P\Psi$ is the projection of the actual wave function Ψ onto a nondegenerate eigenstate (or a group of degenerate eigenstates) of a zero order separable Hamiltonian H_s. H_{eff} is the effective Hamiltonian,[2,3] and Ψ_p is taken to be an eigenstate of H_{eff} with the eigenvalue $\bar{E}$, i.e.:

$$H_{eff}(\bar{E})\Psi_p \equiv \bar{E}\Psi_p . \tag{1c}$$

$\bar{E}$ in Eqs. (1) are the exact eigenvalues of the perturbed Hamiltonian and the WB series for $\bar{E}$ is obtained by an iterative (self-consistent) solution of Eq. (1c). I wish to point out that this arbitrary criterion which has a precise meaning for the WB series is not at all adequate for identifying the time evolution of Ψ_p and as such cannot

serve as an acceptable general means of classification of the molecular eigenstates. The argument goes as follows: If we write

$$H_{eff}(E) \equiv H_0 + R(E) , \qquad (2)$$

where H_0 is a zero-order Hamiltonian, then the time evolution of Ψ_p is given by[2,3]

$$\frac{d}{dt} \Psi_p = -iH_0\Psi_p - \int_0^t d\tau R(t-\tau)\Psi_p(\tau) , \qquad (3a)$$

where

$$R(\tau) = \frac{-1}{2\pi i} \int_{-\infty}^{\infty} dE\, R(E) \exp(-iE\tau) . \qquad (3b)$$

It is clear from Eq. (3) that, in general, the time evolution of Ψ_p is governed by $R(E)$ for all energies E and not only by the local behavior of $R(\overline{E})$. In fact, it is the analytical *global* properties of $R(E)$ in the entire complex E plane which determine the nature of the motion of Ψ_p.[2] Any *local criterion* such as Eq. (1a) which has to do with the behavior of $R(E)$ in a small energy neighborhood around $R(\overline{E})$ cannot therefore predict the time evolution of Ψ_p (since it does not contain enough information). Consequently, condition (1) cannot serve as an adequate criterion for the classification of the nature of Ψ_p. Another way to see this is by considering the following model Hamiltonian describing a single state $|s\rangle$ coupled to a manifold $\{|l\rangle\}$ of levels[4,5]:

$$H = |s\rangle E_s \langle s| + \sum_l |l\rangle E_l \langle l|$$
$$+ \lambda \sum_l \left(V_{sl}|s\rangle\langle l| + V_{ls}|l\rangle\langle s| \right) . \qquad (4)$$

Taking $P \equiv |s\rangle\langle s|$, QHQ is diagonal in this case (where $Q \equiv 1-P$) and the eigenstate of H with energy E is given by

$$|\Psi(E)\rangle = \left[1 + \lambda^2 \sum_l \frac{|V_{sl}|^2}{(E-E_l)^2}\right]^{-1/2} \left[|s\rangle + \lambda \sum_l \frac{V_{ls}}{E-E_l} |l\rangle\right] . \qquad (5)$$

The eigenvalues E are the solutions of the equation:

$$\frac{1}{\lambda^2}(E-E_s) = \sum_l \frac{|V_{sl}|^2}{E-E_l} , \qquad (6)$$

and

$$\eta \equiv \frac{\langle \Psi_p|\Psi_p\rangle}{\langle \Psi|\Psi\rangle} = \left[1 + \lambda^2 \sum_l \frac{|V_{sl}|^2}{(E-E_l)^2}\right]^{-1} . \qquad (7)$$

It is clear by a simple inspection of Eq. (7) that η goes

continuously from 1 (for $\lambda = 0$) to 0 (for $\lambda \to \infty$) and that for some value of λ, η will equal 0.5. The significance of this for the eigenvalue E can be seen by a graphical solution of Eq. (6). The right-hand side, when plotted as a function of E has simple poles for $E = E_l$, $l = 1, 2, \ldots$ and the left-hand side is a straight line with a slope $1/\lambda^2$. For $\lambda \to 0$ (infinite slope), the eigenstates E will be very close to the various E_l. For $\lambda \to \infty$ the solutions E will be bounded between E_l and E_{l+1} (for all l). While the convergence of the WB series will change dramatically as η assumes the value 0.5, it is clear from Eqs. (5)–(7) that nothing dramatic will occur for the eigenvalues E, the eigenstates $|\Psi(E)\rangle$, and the time evolution of the system as η changes from 0.49 to 0.51. What actually happens is that the WB perturbation series "does not know" to which of two (or several) eigenstates of H it should converge. But this is of course a problem of the WB perturbation series, not of the actual eigenvalues and the time evolution of the system! For $\lambda \ll 1$ ($\eta > 0.5$) each eigenvector of H will have a unique "parent" state in H_0 since this is the perturbative (weak coupling) limit. This is the only significance of Eq. (1b). The correlations observed by Hose and Taylor[1] are a simple reflection of the fact that in the perturbative limit, the spectra of H and H_0 are similar. To that end, they could have picked any value of η (not necessarily 0.5) to be the borderline between "small" and "large."

Another problem with the Hose–Taylor analysis is that it is entirely within the "sparse manifold"[6] limit. When the density of states is large enough, we have a quasi-continuum and H_{eff} becomes non-Hermitian with complex eigenvalues.[2,6] The spectra of H and H_{eff} are therefore very different in this case. The most interesting aspect of nonlinear dynamics lies in the transition from the sparse manifold to the statistical (dense manifold) behavior. This issue is not addressed at all by Eq. (1) which applies only to the sparse manifold limit.

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