Detection of a Charge-Density Wave by Angle-Resolved Photoemission

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Angle-resolved photoemission offers a means for determining the presence and the wave vector of an incommensurate charge-density wave in a simple metal.

The purpose of this paper is to show that angle-resolved photoemission offers a means for detecting the presence of an incommensurate charge-density wave (CDW) in a metal. As an example we propose a test for the CDW model of potassium.[1,2] The abundance of experimental evidence supporting the hypothesis of a CDW ground state in metallic potassium has recently been reviewed.[3]

The most conclusive evidence for the presence of a CDW would be a diffraction "picture" of the satellite spots associated with the wave vector $\vec{Q}$ of the CDW. In potassium such spots have not been seen.[4] However, the surmised location of the spots is very close to the [110] Bragg reflection. Furthermore, a large, expected phason temperature factor[2] may make them nearly unobservable.[5] We show that angle-resolved photoemission offers an alternative means for detecting the presence of a CDW and can be used to determine the CDW wave vector $\vec{Q}$.

Potassium films grown on smooth, amorphous substrates are known to have a [110] direction normal to the surface.[6] (This should, of course, be checked in each experiment.) In addition, there is evidence that the direction of $\vec{Q}$ in such a film is along that [110] direction which is normal to the surface.[7] For this case the geometry is especially simple, and a measurement of the energy distribution of electrons emitted *normal* to the surface can detect the presence of a CDW.

Figure 1 illustrates the process of photoemission. An incoming photon with energy $\hbar\omega$ excites an electron from an initial state E_i (measured from the Fermi energy E_F) to a final state. The work function of the metal is φ. The electron escapes from the metal with kinetic energy K. Knowledge of K and $\hbar\omega$ allows one to determine the energy of the initial state,

$$E_i = K + \varphi - \hbar\omega . \tag{1}$$

The energy of the final state is $E_i + \hbar\omega$. Typically, angle-resolved electron-energy distribution curves (AREDC's) are displayed by plotting the spectral intensity of emitted electrons versus E_i.

AREDC's give information about the energy and direction of electrons emitted from the sample. In the case under consideration electrons emitted normal to the surface carry information about the energy bands in the [110] direction. For this discussion we shall consider only electrons emitted from states in the bulk (and shall ignore complications arising from electrons in surface states).

As is well known, there is no absorption of photons in a free-electron gas since momentum and energy cannot be conserved. In the presence of the periodic potential of a crystal, *or a CDW*, transitions are allowed. For the case being considered, in which electrons are emitted along [110], it is sufficient to examine the one-electron potential

$$V(\vec{r}) = v_{110}\cos\vec{G}_{110}\cdot\vec{r} + v\cos\vec{Q}\cdot\vec{r} . \tag{2}$$

The first term is the [110] component of the crystal potential, and the second term is the CDW potential. We neglect here other components of the crystal potential since their magnitude is much smaller. Thus, we neglect the secondary-cone emission of Mahan.[8]

In the nearly-free-electron model the effect of $V(\vec{r})$ is to mix the plane-wave state $\vec{k}\pm\vec{G}_{110}$ and $\vec{k}\pm\vec{Q}$. The amount of mixing can be determined by perturbation theory. With these perturbed states there can be absorption as illustrated in

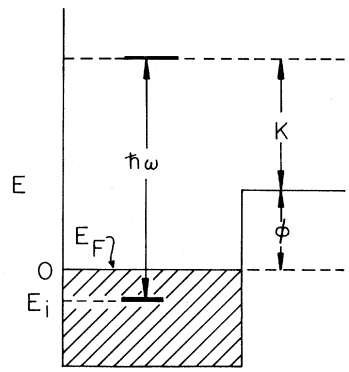

FIG. 1. Sketch of photoemission. A photon with energy $\hbar\omega$ excites an electron in an initial state with energy E_i below the Fermi energy E_F. The work function of the metal is φ. The emitted electron has kinetic energy K.

Fig. 2. Because of the multiple periodicity of $V(\vec{r})$ it is easiest to think in the extended-zone scheme. If the CDW is absent (there is no gap at point A and) the only transitions allowed are between states separated in k space by $\vec{G}_{110}$ (such as A to C). For $\hbar\omega$ greater than the difference in energy between points C and A, there can be no transitions since A is at the Fermi surface in the [110] direction. If a CDW is present, additional transitions are possible (such as A to B) between states separated by $\vec{Q}$ in k space. There are no "Q" transitions for $\hbar\omega$ greater than the energy difference between A and B. As we show below, by varying $\hbar\omega$ it is possible to discriminate between "G" and "Q" transitions and to determine the difference $G_{110}-Q$.

AREDC's for electrons emitted normal to the surface are shown in Fig. 3 for a number of photon energies. The numerical values, appropriate to potassium, are based on nearly-free-electron energies with $V_{110}=0.4$ eV,[9] a CDW energy gap $V=0.6$ eV,[1] and a Fermi surface in critical contact[1] with the CDW energy gaps.

The photon energies in Fig. 3 are only meant to be illustrative because of uncertainties in band structure and many-body effects not included in the energies of the electron states. Therefore we have added an unknown energy η to all photon energies. (η would be zero for the nearly-free-electron model under discussion.) The experiment we propose here would be of interest even if the only outcome were a determination of η.

Because of the uncertainties related to band structure and many-body effects we do not estimate peak heights or widths. Since v_{110} and V are of comparable size, the relative heights of the G and Q transitions are expected to be comparable. We choose to represent the distribution of emitted electrons by Gaussians of equal height and full width ~0.3 eV. We arbitrarily truncate a Gaussian when it would lead to spectral intensity above $E_i=0$.

We start first with a high photon energy and construct a series of curves for decreasing values of $\hbar\omega$. As discussed above, for $\hbar\omega$ greater than the energy difference between points A and C in Fig. 2, there will be no transitions. Such a case (where there are no external photoelectrons) is shown in the top curve of Fig. 3.

As $\hbar\omega$ is reduced, the G transition first be-

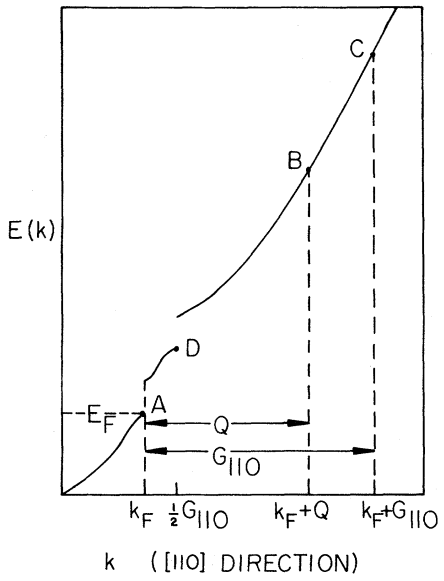

FIG. 2. Energy versus wave vector in the [110] direction for a nearly-free-electron metal containing a CDW. G_{110} is the [110] reciprocal lattice vector and Q is the CDW wave vector. The CDW energy gap is at A and the Brillouin zone gap is at D. With no CDW present only transitions between states separated by G_{110} are allowed, e.g., between points A and C. The periodicity Q of the CDW allows additional transitions such as from A to B.

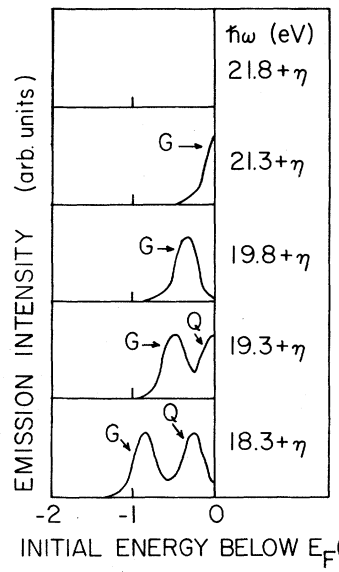

FIG. 3. A sketch of intensity of emitted electrons versus energy of the initial state below the Fermi energy for a number of photon energies. η is a parameter to account for band-structure and many-electron effects. It is zero for the nearly-free-electron model. Peaks labeled G are allowed by the crystal potential, and peaks labeled Q are allowed by the CDW potential. See text for details.

comes possible for $\hbar\omega = 21.5$ eV, an excitation from A to C in Fig. 2 (with the initial state A at the Fermi energy). This is illustrated in the second curve of Fig. 3 for an $\hbar\omega$ slightly below threshold. For smaller $\hbar\omega$ the initial state for the G transition moves to lower energy as shown in the third curve of Fig. 3.

Below $\hbar\omega = 19.5$ eV the Q transition (A to B in Fig. 2) becomes possible as shown in the fourth curve of Fig. 3. As $\hbar\omega$ is decreased further, the energies of initial states for both G and Q transitions decrease. When the Q transition is first allowed, the center of the G peak is ~0.4 eV below the Fermi energy in the nearly-free-electron model. The difference in initial energies for the two transitions increases to ~0.6 eV as $\hbar\omega$ is lowered.

The calculation is based on a value $Q = 1.33(2\pi/a)$. The separation between the peaks can be used to determine the difference $G_{110} - Q$. [This depends on a knowledge of $E(\vec{k})$ for the initial- and final-state bands.] Thus, angle-resolved photoemission can be used both to detect the presence of a CDW and to measure its periodicity.

We have performed a similar analysis for sodium. Since the Fermi energy in sodium is about 1.5 times that in potassium, the G transition (A to C in Fig. 2) occurs for $\hbar\omega = 33.4$ eV in the nearly-free-electron model. The Q transition (A to B in Fig. 2) occurs for $\hbar\omega = 31.4$ eV. The spacing between the two peaks is similar to that in potassium, when a value $Q = 1.35(2\pi/a)$ is assumed for sodium. However, because V_{110} in Na is only ~0.2 eV,[9] and the CDW energy gap V is thought to be about 1.2 eV, the peak height for the G transition may be much smaller than that for the Q transition. This disparity could lead to experimental difficulties.

We have shown that angle-resolved photoemission offers a means to detect the presence of an incommensurate CDW. The crucial idea is that the periodicity $\vec{Q}$ of the CDW permits new transitions compared to those allowed by the periodicity of the crystal. By plotting AREDC's for various incident photon energies, it should be possible to identify any additional transitions and (if present) to determine $\vec{Q}$.

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Replica-Symmetry Breaking in Spin-Glass Theories

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The instability which arises in current mean-field theories of spin-glasses is removed, within the replica method, by breaking the symmetry between replicas. For the Sherrington-Kirkpatrick (SK) model the spin-glass order parameter has the expansion $q = t + t^2 + O(t^3)$ ($t = 1 - T/T_c$), which differs at $O(t^2)$ from that given by SK.

Before substantial progress can be made in understanding any phase transition it is essential to obtain a sound understanding of the appropriate mean-field theory. For the spin-glass phase transition the model of Sherrington and Kirkpatrick[1] (SK) provides a suitable starting point. This is a model of Ising spins coupled by random, infinite-ranged exchange interactions independently distributed with a Gaussian probability density. The disorder is quenched, and so the free energy, rather than the partition function, must be averaged over the bond distribution. SK attempted to solve this model, using the "n-replica method,[2]" by means of which one writes $\ln Z = \lim_{n\to 0}(Z^n - 1)/n$ and recognizes that Z^n is the partition function for n identical replicas of the original system. The bond averages may now be taken at the outset yielding a translationally invariant model of coupled replicas. This may be solved in the thermodynamic limit and the analy-