moved if impurities whose separation is smaller than a critical value cannot contribute to the Raman spectrum. A possible explanation for this feature is that an exciton interacting equally strongly with more than one impurity cannot be bound and serve as necessary intermediate state. Using the same parameters as above, i.e.,  $R_{\rm exc}$ = 40 Å and  $\alpha^{-1}$  = 20 Å, the critical separation would be  $R_c \sim 120$  Å; although this estimate is crude, a more accurate determination requires the study of the complex interaction of three neutral entities.

In conclusion we propose further Raman studies of spin-flip transitions at lower magnetic fields to check the detailed field dependence of the multi-spin-flip peaks. It is evident from the discussion following Eq. (1) that the higher-order peaks  $\Delta S = 3, 4, \cdots$  may become observable at sufficiently low fields, and exhibit a strong field dependence. Note, however, that our predictions require low temperatures such that  $k_{\rm B}T < g\mu H$ . Since the parameters in our theory are very sensitive to the form of the wave functions, the multiple spin-flip experiments may provide a check on future calculations on excitons bound to neutral impurities.

We are indebted to J. F. Scott for stimulating our interest in this problem, and to W. F. Brinkman for valuable comments.

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<sup>1</sup>J. J. Hopfield and D. G. Thomas, *Light Scattering Spectra of Solids* edited by G. Wright (Springer, Berlin, 1969), p. 255, and Phys. Rev. 175, 1021 (1968).

<sup>2</sup>J. F. Scott and T. C. Damen, preceding Letter [Phys. Rev. Lett. 29, 107 (1972)].

<sup>3</sup>For convenience we consider the field direction as the axis of spin quantization.

<sup>4</sup>This is the basic Compton scattering process. See, for example, W. Heitler, *Quantum Theory of Radiation* (Clarendon Press, Oxford, 1936), p. 132; J. J. Sakurai, *Advanced Quantum Mechanics* (Addison-Wesley, Reading, Mass., 1967), p. 215.

<sup>b</sup>L. D. Landau and E. M. Lifshitz, *Quantum Mechanics* (Addison-Wesley, Reading, Mass., 1958), p. 150.

<sup>6</sup>Y. Toyozawa, J. Phys. Soc. Jap. <u>17</u>, 986 (1962).

<sup>7</sup>R. M. Martin, in *Proceedings of the Second International Conference on Light Scattering in Solids*, edited by M. Balkanski (Flammarion, Paris, 1972), p. 25.

<sup>8</sup>E. I. Rashba and G. E. Gurgenishvili, Fiz. Tverd. Tela <u>4</u>, 1029 (1962) [Sov. Phys. Solid State <u>4</u>, 759 (1962)].

<sup>9</sup>A crude estimate, using the analogy with the neutral hydrogen molecule and taking into account the dielectric constant and electronic effective mass appropriate to CdS, gives  $R_{\rm exc} \gtrsim 35$  Å.

## Gor'kov-Eliashberg Effect in One-Dimensional Metals?

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We point out the possibility that the potentially important dielectric effect first considered by Gor'kov and Eliashberg for minute metallic particles may well occur in the recently identified one-dimensional metals formed by the mixed-valency planar complex compounds of Pt. Our idea is conditional on the validity of a recently proposed model of interrupted metallic strands.

Following the original work of Kubo<sup>1</sup> on the electronic properties of small metallic particles, Gor'kov and Eliashberg<sup>2</sup> (GE) made in 1965 the most remarkable theoretical prediction that the electronic polarizability of a metallic particle —sufficiently minute that its electronic energy levels are discrete—should be enormously enhanced with respect to the classical polarizability which one would have expected on the basis of elementary electrostatics. The effect should have been observable in electric fields and at ab-

solute temperatures such that the discrete electronic energy levels of the minute particle remain unmixed, i.e., under the physical conditions for which the minute-metallic-particle system could be regarded as a dielectric insulator. Besides the considerable intrinsic physical interest of the effect, such an anomalous enhancement of the electronic polarizability of a minute metallic particle would have important technological application, a fact fully realized by GE.<sup>2</sup> Unfortunately, however, several recent experimental investigations<sup>3</sup> of the effect have confirmed that it does not arise. As subsequently discussed by Strässler, Rice, and Wyder<sup>4</sup> the explanation of these negative findings is quite simple. Although GE correctly recognized that the electronic susceptibility of a macroscopic system of discrete electronic energy levels would be exceedingly large, its effect on the polarizability of a minute metallic particle will not be observable because of the canceling effect of the depolarization field of the particle's finite geometry.

It is the purpose of the present Letter to point out the intriguing possibility that there may exist a class of metallic systems for which the negative effect of a depolarization field will not arise, and, hence, for which an anomalous, and potentially useful, dielectric effect could be expected. The particular metallic systems we have in mind are the recently identified<sup>5,6</sup> quasi-one-dimensional (1D) metals formed by the mixed-valency planar complex compounds<sup>7</sup> (MVPC) of Pt. As will be seen our idea is conditional on the validity of the so-called "interrupted-strand model" that has been advanced<sup>5,8</sup> to interpret the observed electronic properties of these compounds. The interrupted-strand model simply views<sup>9</sup> the real crystalline compound to consist of a system of collinear metallic strands, each of which is interrupted by a series of perfectly insulating lattice defects. A small distribution of such defects replaces the defect-free strand by a linear sequence of 1D "metallic boxes." The similarity with the minute-metallic-particle problem becomes evident when, on accepting the interruptedstrand model, one realizes that the spacing of the electronic energy levels of a typical 1D box will be large for practically occuring concentrations of residual lattice defects (e.g., 1 at.%). If we denote by  $l_0$  the average length of a metallic box, the pertinent spacing  $\Delta$  of the energy levels will be just<sup>2,10</sup>  $\Delta = \hbar \pi v_F / l_0$ , where  $v_F$  denotes the 1D Fermi velocity of the ideal strand. With  $l_0 \sim 300$ Å, corresponding to an uninterrupted chain sequence of ~100 Pt atoms and a typical Pt-Pt distance  $a \simeq 3$  Å, we have  $\Delta \sim 0.07$  eV ( $\Delta/k_B \sim 800^{\circ}$ K) for  $v_{\rm F} \sim 10^8$  cm sec<sup>-1</sup>. The 1D metallic box of the interrupted-strand system may thus be regarded as the analog of the minute metallic particle considered by GE. For the 1D box, however, the difficulty associated with a depolarization field should not arise.

We present below an explicit model calculation which predicts the system of interrupted strands to exhibit a giant dielectric constant  $\epsilon$  along the strand axis, given by

$$\epsilon \simeq \frac{1}{2} (q_s l_0)^2, \tag{1}$$

where  $q_s$  denotes the Fermi-Thomas screening wave vector of the conduction electrons. Since for metallic densities  $q_s \sim a^{-1}$ , Eq. (1) shows that, order-of-magnitude-wise,  $\epsilon$  will be given by the square of the number of Pt atoms comprising the mean metallic box. From the practical standpoint, this suggests the important possibility of synthesizing large controllable dielectric constants by manipulation of the defect concentration of the metallic strands. Moreover, since the characteristic relaxation time  $\tau_R$  of the polarization process involved in the present effect can be expected to be very short,<sup>8</sup> i.e.,  $\tau_R \sim l_0/$  $v_{\rm F}$ , it is not inconceivable that the MVPC compounds of Pt could constitute a new class of highfrequency high-dielectric-constant materials.

At absolute zero the available conduction electrons of the interrupted strand system will be distributed over the allowed stationary energy levels  $\epsilon_{n,l}$  of the various boxes of the interrupted-strand system in such a way that all levels  $< \epsilon_{\rm F}$  are occupied and all those  $> \epsilon_{\rm F}$  are empty. In the present calculation we will assume that  $\epsilon_{n,l}$  is of the form<sup>10</sup>

$$\epsilon_{n,l} = \epsilon_k(v_p; l/a) \quad (k = n\pi/l; \ n = 1, 2, \cdots), \tag{2}$$

where  $v_p$  denotes the intrinsic periodic potential, and *a* the lattice spacing, of the ideal strand. The appearance of the ratio l/a in the argument of  $\epsilon_k$  indicates the influence of the box boundaries on  $\epsilon_{n,l}$ . If we denote by  $\psi_{n,l}(x)$  the eigenfunction corresponding to  $\epsilon_{n,l}$ , it follows from linear-response theory<sup>11</sup> that the static dielectric constant  $\epsilon$  of the strand system in a direction parallel to a strand axis is

$$\epsilon = 1 + \frac{4\pi n_A e^2}{L} \sum_{n,n'} \sum_{l} |\langle n, l | x | n', l \rangle|^2 \frac{f_{n,l} - f_{n',l}}{\epsilon_{n',l} - \epsilon_{n,l}}, \quad (3)$$

where  $n_A$  denotes the number of strands per unit area in a plane perpendicular to the strand axis, L the macroscopic length of the system,  $f_{n,l}$  the Fermi-Dirac occupation of the level  $\epsilon_{n,l}$ , and

$$\langle n, l | x | n', l \rangle = \int_0^l \psi_{n, l} *(x) x \psi_{n', l}(x) \, dx.$$
(4)

In view of the sum over the different box lengths l and the dependence of  $\epsilon_{n,l}$  on l, (3) is a much more complicated expression than a similar expression derived by GE for a minute metallic sphere. In order to evaluate it we shall first assume that the mean box length  $l_0$  is sufficiently large (say  $l_0/a > 10$ ) that we may always neglect

the dependence of  $\epsilon_{n,l}$  on the nature of the box boundaries, i.e., we shall take  $\epsilon_{n,l} = \epsilon_k (v_p; \infty)$  $\equiv \epsilon_k$ , in which case  $\epsilon_{n,l}$  depends on l only through its dependence on k. Secondly, we shall assume that the  $\psi_{n,l}$  have the form  $\psi_{n,l} = (2/l)^{1/2} \sin(kx)$ , so that the diagonal matrix elements in (4) are just l/2 for all n. Also, since the squares of the nondiagonal matrix elements will be small by comparison to the square of the diagonal matrix element we shall, as a third approximation, omit them from the sum in (3). The dielectric constant thus becomes

$$\epsilon = 1 + \frac{4\pi n_A e^2}{L} \sum_{l} \left(\frac{l}{2}\right)^2 \sum_{n=1}^{\infty} \delta(\epsilon_{n,l} - \epsilon_F)$$
 (5)

$$=1+\frac{2\pi n_A e^2}{L\hbar v_F}\sum_l l^2 \sum_{n=1}^{\infty} \delta\left(\frac{n\pi}{l}-k_F\right), \qquad (6)$$

where we have written

$$(f_{n,l} - f_{n',l}) / (\epsilon_{n',l} - \epsilon_{n,l}) = -\partial f_{n,l} / \partial \epsilon_{n,l}$$
$$= \delta(\epsilon_{n,l} - \epsilon_{\rm F})$$

and introduced the limiting Fermi wave vector  $k_{\rm F}$  and Fermi velocity  $v_{\rm F} = \hbar^{-1} (\partial \epsilon_k / \partial k)_{k_{\rm F}}$ . In going from (5) to (6) we have introduced an additional factor of 2 to account for a spin- $\frac{1}{2}$  degeneracy for each energy level. Our final approximation is to assume that the number  $\delta N$  of boxes that can be found with a length between l and  $l + \delta l$  is given by the distribution law

$$\delta N = N_d \exp(-l/l_0) \, \delta l/l_0 \quad (l_0 = L/N_d), \tag{7}$$

where  $N_d$  denotes the number of lattice defects in a strand. Equation (7) is actually exact in the limit of a dilute random distribution of defects.<sup>10</sup> With Eq. (7) the sum in Eq. (6) over l may be replaced by an integral, which, because of the presence of the  $\delta$  function in Eq. (6), may be readily evaluated to give

$$\epsilon = 1 + \frac{2\pi n_A e^2 l_0}{\hbar v_F k_F} \sum_{n=1}^{\infty} \left(\frac{n\pi}{k_F l_0}\right)^3 \exp(-n\pi/k_F l_0).$$

The sum over *n* may be evaluated exactly, and for  $l_0 k_F \gg 1$ , which is the limit of interest to us, it is  $6k_F l_0/\pi$ . Thus we obtain

$$\epsilon - 1 = \frac{12(n_A e^2 l_0^2)}{\hbar v_F} = \frac{1}{2}(q_s l_0)^2 \tag{8}$$

which, since  $q_s l_0 \gg 1$ , is the result (1). We have defined a formal Fermi-Thomas screening wave vector as  $q_s^2 = 24n_A e^2/\hbar v_F$ . Equation (8) may be compared with the result  $4\pi\chi \sim r^2 q_s^2$  found by GE for the electronic susceptibility  $\chi$  of a minute metallic sphere of radius r.<sup>11</sup>

In the interrupted-strand model the system of collinear metallic strands is considered to behave as a dielectric insulator in dc fields at absolute zero.<sup>8,10</sup> The finite dc conductivities observed for the MVPC compounds at  $T \neq 0$  are imagined<sup>5,10</sup> to result from the thermally excited tunneling of an electron from one metallic box to either an adjacent or distant one. The existence of this finite dc conductivity will hinder the possible observation at normal temperatures of the above predicted giant dielectric constant. However, it should be readily observed at low temperatures where the thermally activated dc conductivities will be negligible. Berenblyum  $et \ al^{12}$  have, in fact, already reported a provisional giant dielectric constant  $\sim 10^3$  for the MVSP compound  $K_2Pt(CN)_4Br_{0.3}(H_2O)_{2.3}$  at microwave frequencies  $(\omega \tau_R \sim 10^4)$  and liquid-helium temperatures. If we attribute this observed dielectric constant to that given by (8), we obtain the estimate<sup>13</sup>  $l_0 \sim 100$ Å, on using the value  $v_{\rm F} \simeq 10^8 {\rm ~cm~sec^{-1}} {\rm ~deduced^8}$ from independent measurements<sup>5,6</sup> of the plasma frequency.

Clearly, further experimental studies of the low-temperature dielectric constant of the MVSP compounds would be of great interest in the context of the suggestions of this Letter.

We have enjoyed many stimulating discussions with D. Kuse, S. Strässler, and H. R. Zeller.

<sup>1</sup>R. Kubo, J. Phys. Soc. Jap. <u>17</u>, 975 (1962).

<sup>2</sup>L. P. Gor'kov and G. M. Eliashberg, Zh. Eksp. Teor. Fiz. <u>48</u>, 1407 (1965) [Sov. Phys. JETP <u>21</u>, 940 (1965)].

<sup>3</sup>R. Dupree and M. A. Smithard, J. Phys. C: Proc. Phys. Soc., London <u>5</u>, 408 (1972); F. Meier and P. Wyder, Phys. Lett. <u>39A</u>, 51 (1972).

<sup>4</sup>S. Strässler, M. J. Rice, and P. Wyder, Phys. Rev. B (to be published); R. Kubo, Comments Solid State Phys. <u>1</u>, 61 (1968), has discussed a possible difficulty with the Gor'kov-Eliashberg effect that arises from considerations of the oscillator strength sum rule.

<sup>5</sup>D. Kuse and H. R. Zeller, Phys. Rev. Lett. <u>27</u>, 1060 (1971); see also, H. R. Zeller, Phys. Rev. Lett. <u>28</u>, 1452 (1972).

<sup>6</sup>H. P. Geserich *et al.*, Phys. Status Solidi (a) <u>9</u>, 187 (1972).

<sup>7</sup>For a review see K. Krogmann, Angew. Chem., Int. Ed. Engl. <u>8</u>, 35 (1969); also, M. J. Minot and J. H. Perlstein, Phys. Rev. Lett. <u>26</u>, 371 (1971).

<sup>8</sup>M. J. Rice and J. Bernasconi, Phys. Lett. <u>38A</u>, 277 (1972), and to be published.

<sup>9</sup>In view of the recent papers by A. N. Bloch, R. B. Weisman, and C. M. Varme [Phys. Rev. Lett. <u>28</u>, 753 (1972)] and by J. H. Perlstein *et al.* (to be published) we should point out the recent observation [M. J. Rice, Phys. Lett. <u>39A</u>, 289 (1972)] to the effect that the presence of a weak random potential along the strands in the real MVPC compounds will by no means rule out the existence of a 1D metallic state, at least, in the sense of the interrupted-strand model.

<sup>10</sup>A detailed microscopic development of the interrupted-strand model has been recently completed [M. J. Rice and J. Bernasconi, to be published]. <sup>11</sup>See, for example, Ref. 3.

<sup>12</sup>A. S. Berenblyum *et al.*, Pis'ma Zh. Eksp. Teor. Fiz. <u>13</u>, 619 (1971) [JETP Lett. <u>13</u>, 440 (1971)].

<sup>13</sup>A similar estimate was found in Ref. 7 in which a simple phenomenological model of the optical properties of a 1D metal, based on the interrupted-strand model, was presented.

## Time-Differential Quadrupole Interaction of Cd<sup>111</sup> Nuclei Implanted by ( $\alpha$ , 2n) Reactions into a Cubic Ag Lattice\*

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The quadrupole interaction of  $Cd^{111}$  ions following the decay of  $In^{111}$  implanted by  $(\alpha, 2n)$  reactions into a cubic (fcc) Ag lattice has been measured with the time-differential perturbed angular correlation method. In addition to a smeared-out quadrupole interaction with a centroid frequency of  $\overline{\omega}_Q \approx 2.5$  MHz, about 10% of the implanted  $Cd^{111}$  ions experience a unique quadrupole interaction with  $\omega_Q^0 = 115$ , 130, or 140 MHz, depending on the primary  $\alpha$  beam. Annealing causes the quadrupole perturbation to vanish completely.

In a recent Letter, McDonald, Lesser, and Fossan<sup>1</sup> reported measurements of the quadrupole interaction of the 247-keV state of Cd<sup>111</sup> implanted by recoil in Coulomb excitation into polycrystalline Cd metal. The results of these measurements indicate that most of the recoiling Cd<sup>111</sup> ions come to rest at the regular lattice sites of the polycrystalline Cd catcher and that the quadrupole interaction is the same as the lattice quadrupole interaction in the hexagonal Cd metal which has been observed by perturbed angular correlation methods.<sup>2</sup> No evidence was found of the recoiling ions ending up interstitially or of crystal damage such as nearby vacancies or interstitials in the host lattice produced by the recoiling ion or the primary particle beam.

The present work describes the observation of a nonvanishing static quadrupole interaction of  $In^{111}-Cd^{111}$  ions that were implanted into a cubic (fcc) Ag lattice by the recoil experienced in an  $(\alpha, 2n)$  reaction. Since the static quadrupole interaction of ions at regular lattice sites in a nondamaged cubic lattice would vanish, the experiments clearly indicate the presence of interstitial occupations by the Cd<sup>111</sup> ions or, more likely, of considerable damage of the cubic Ag host lattice.

The experiments consisted of bombarding polycrystalline Ag foils at room temperature with 22-MeV  $\alpha$  particles from the Purdue tandem accelerator and with 35- and 45-MeV  $\alpha$  particles from the Argonne cyclotron, thus producing, by an  $(\alpha, 2n)$  reaction, In<sup>111</sup> nuclei in the Ag lattice with recoil energies in the MeV range. The In<sup>111</sup> ions slow down, mainly by electronic interactions. Nuclear stopping becomes important at the end of the slowing-down process and host atoms are displaced from their lattice sites by nuclear collisions. After the In<sup>111</sup> ions come to rest somewhere in the cubic Ag lattice, they decay by electron capture with a half-life of 2.8 days to the 419-keV excited state of Cd<sup>111</sup>. The 172-keV  $\gamma$ radiation de-exciting this state populates a state of 247-keV excitation energy which has a lifetime of  $\tau = 123$  nsec before it decays to the Cd<sup>111</sup> ground state. During this lifetime the 247-keV,  $I = \frac{5}{2}$ Cd<sup>111</sup> state is exposed to hyperfine fields which can be accurately measured by observing the time dependence of the perturbation of the angular correlation between the 172- and 247-keV  $\gamma$ rays of Cd<sup>111</sup>.<sup>3</sup>

It is well known that the atomic shell of the  $Cd^{111}$  ion recovers within a short time (<  $10^{-11}$  sec) from the preceding electron capture in a metallic environment.<sup>3</sup> Time-dependent electric field gradients due to vacancy diffusion may produce small perturbation. Within the time range of interest here (100 nsec), and since the relaxation times are of the order of several  $\mu$ sec at room temperature,<sup>4</sup> these perturbations can be neglected. Magnetic hyperfine interactions can be excluded since both Cd and Ag are diamagnetic. Consequently only the interaction of the electric