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Anomalous magnetoresistance near the superconductor-insulator transition in ultrathin films of a-Mo_xSi_{1-x}

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We have made systematic studies for both the zero-field and field-driven superconductor-insulator transitions in a series of 4-nm-thick films of amorphous Mo_xSi_{1-x} at temperatures T down to ~0.05 K and fields B up to 15 T. For superconducting films, we have observed an anomalous peak in the magnetoresistance $R(B)$ and a subsequent decrease in $R(B)$ with increasing B at low temperatures in fields higher than the critical field B_{xxC}. This result, together with finding that the magnetoresistance is always positive for insulating films, may suggest the presence of localized Cooper pairs even at $B>B_{xxC}$ in the limit of $T\rightarrow 0$.

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The superconductor-insulator (S-I) transition in two dimensions (2D) has attracted intense recent attention from the viewpoint of quantum phase transitions, which occur at zero temperature $(T=0)$.[1] Despite numerous theoretical[1–3] and experimental[4–19] effort, there still remain some basic and interesting problems to be settled. In particular, the existence of the universal critical sheet resistance R_c for the S-I transition and that of the Bose-insulator phase[6,8,12] are most interesting problems which have been actively discussed. The amorphous $(a$-) Mo_xSi_{1-x} films[15] used in this work are very suitable in studying the 2D S-I transition in the following respects: First, ultrathin (4-nm) and microscopically disordered amorphous films which undergo a superconducting transition can be obtained. Judging from the sharp (resistive) superconducting transition curves observed down to our experimental resolutions $R(T)\sim 10^{-2}\ \Omega$, the uniformity of our films is considered to be fairly good. Second, disorder [more precisely, a resistance in the normal state $R_n(10\ \mathrm{K})$ at $T=10$ K] of the films together with the superconducting transition temperature can be continuously changed by varying the Mo concentration x for fixed thickness.

Using a series of 4-nm-thick films of a-Mo_xSi_{1-x}, we recently performed systematic studies for both the zero-field (disorder-driven) and field-driven S-I transitions at $T>0.5$ K in fields B of up to 7 T.[15] For superconducting films, a Kosterlitz-Thouless transition temperature T_{KT} determined from the current (I)-voltage (V) characteristics was very close to the mean-field transition temperature T_{C0}; $\Delta T=T_{C0}-T_{\mathrm{KT}}$ was typically ~0.01 K. Both T_{KT} (T_{C0}) and the critical field B_{xxC} for the field-driven S-I transition decreased monotonically with increasing $R_n(10\ \mathrm{K})$ and vanished at around $R_n(10\ \mathrm{K})\sim 2\ \mathrm{k}\Omega$. Critical resistances for the zero-field and field-driven S-I transitions were found to be nonuniversal and apparently smaller than $h/4e^2\sim 6.45\ \mathrm{k}\Omega$. These results seem to be inconsistent with the ''dirty boson'' model,[1] in which the insulating state near the S-I transition is dominated by the properties of localized Cooper pairs and the S-I transition is a $T=0$ quantum phase transition with a quantum critical point at which $R\sim h/4e^2$.

In this paper we present the transport properties for the same Mo_xSi_{1-x} system at even lower temperatures down to ~0.05 K and higher fields up to ~15 T. We have found that the results mentioned above stay essentially unchanged. However, for superconducting films a peculiar peak in the magnetoresistance has been observed at fields higher than the critical field B_{xxC}. Such anomalous behavior is only visible at low enough temperatures $(T<0.2\ \mathrm{K})$. Combined with the results for insulating films, we will suggest the possibility that localized Cooper pairs may be present even on the insulating side of the field-driven S-I transition $(B>B_{xxC})$ in the limit of $T\rightarrow 0$.

Mo_xSi_{1-x} films, 4 nm thick and 0.4 mm wide, were prepared by coevaporation of pure Mo and Si in the pressure better than 10^{-8} Torr. We employed a gradient deposition technique to obtain a series of samples with continuously changing x. The structure of the films used in this study $(x=26-61$ at. %) was confirmed to be amorphous by means of transmission electron microscopy (TEM), consistent with the past reported results for 100-nm-thick films.[20] For measurements above ~0.5 K the films were directly immersed into liquid ^{3}He or ^{4}He, while for measurements down to ~0.05 K they were attached to the cold plate of the mixing chamber in our dilution refrigerator. The resistance was measured by standard four-terminal dc and ac locking methods with a current $I\sim 10$ nA. For insulating films, we measured the I-V characteristics at $T\sim 0.05$ K and checked Ohmic behavior at a measurement current. In studying the resistive transition region for superconducting films, we also measured I-V characteristics as a function of T. The field was directed perpendicularly to the film surface. For measurements at $B=0$, we applied a small perpendicular field $(\approx 9\times 10^{-5}\ \mathrm{T})$ to cancel the ambient field.[18]

Figure 1(a) illustrates the temperature dependence of the sheet resistance $R(T)$ in $B=0$ for ten selected films. Films with $R_n(10\ \mathrm{K})$ smaller than 1.8 kΩ (films 1–4) achieve global superconductivity, while those with $R_n(10\ \mathrm{K})$ larger than 2.5 kΩ (films 5–10) behave like an insulator, showing an increase in R at low temperatures. Here it is implicitly assumed that there are only two states in 2D at $T\rightarrow 0$, except right at the S-I transition. Thus, the critical resistance $R_{nC}(10\ \mathrm{K})$ for the zero-field S-I transition is ~2 kΩ, consistent with the previous results at $T>0.5$ K.[15] Except for the reduced values of the critical resistance, these data are remarkably similar to the past reported data for amorphous thin films

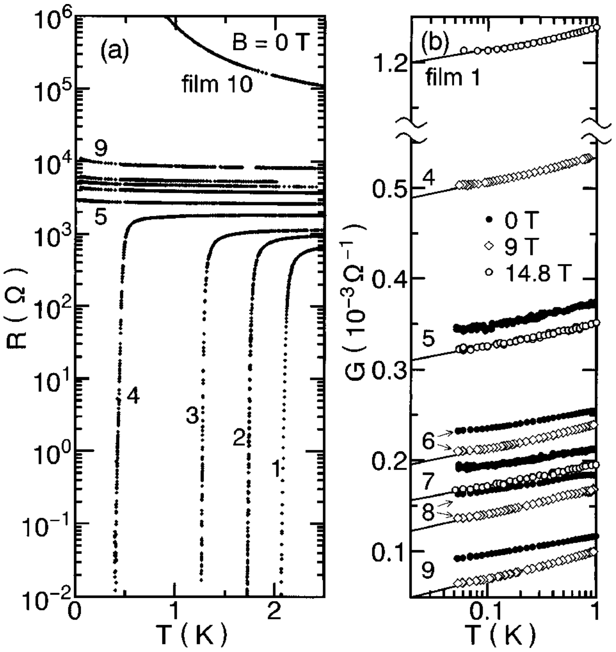

FIG. 1. (a) The sheet resistance R vs temperature T in $B=0$ for a series of 4-nm-thick $\mathrm{Mo}_x\mathrm{Si}_{1-x}$ films with varying x; $x=0.61$ (film 1)–0.26 (film 10). (b) The sheet conductance $G(=1/R)$ for seven selected films in $B=0$ and high fields (9 or 14.8 T) vs ln T. The straight lines are fits to the points.

prepared by quench condensation.[4] Figure 1(b) shows the sheet conductance $G(T)=1/R(T)$ at high fields ($B=9$ or 14.8 T) vs ln T for seven selected films. These fields are high enough to suppress the superconductivity in films 1–4. Except for the most resistive film (film 10), where $G(T)$ is nearly the variable-range-hopping type, the temperature variation of $G(T)$ for the remaining films is well reproduced by the formula expected in the 2D weak-localization theory: $G(T)=G_0+p(e^2/2\pi^2\hbar)\ln T$, where G_0 and p are constants independent of T; the values of p stay in the limited range between 0.8 and 1.0 for these films. This result indicates that not only superconducting films but also insulating films (films 5–9) lie in the weakly localized regime for fermions. This is in contrast to the previous measurements for a series of ultrathin Pb films.[16]

In Fig. 2 $R(T)$ for film 4 in various fields ($B=0-9$ T) is typically plotted. With increasing B, the slope of the isomagnetic curve below ~0.5 K changes its sign from positive to negative at around $B=2.2-2.5$ T, indicating that a field-induced transition from a state where R is zero to a state where R is infinity takes place at $T=0$. This behavior is different from what has been reported for the similar 2D system of a-$\mathrm{Mo}_{43}\mathrm{Ge}_{57}$ films,[17] where no superconducting vortex-glass state exists even at $T=0$. From the R vs B plots at fixed T ($\geqslant 0.05$ K), the critical field B_{xxC} and critical resistance $R_C(B=B_{xxC})$ for the field-driven S-I transition are clearly estimated to be ≈2.3 T and ≈1.8 kΩ, respectively. We have found that $R_C(B=B_{xxC})$ is sample dependent and approximately equal to $R_n(10\text{ K})$ in the range $R_n(10\text{ K})=0.77-1.8$ kΩ. This result, as well as $R_{nC}(10\text{ K})\sim 2$ kΩ (for the zero-field S-I transition), indicates the apparent lack of universality for the critical resistances.

It is interesting to note that as B increases further

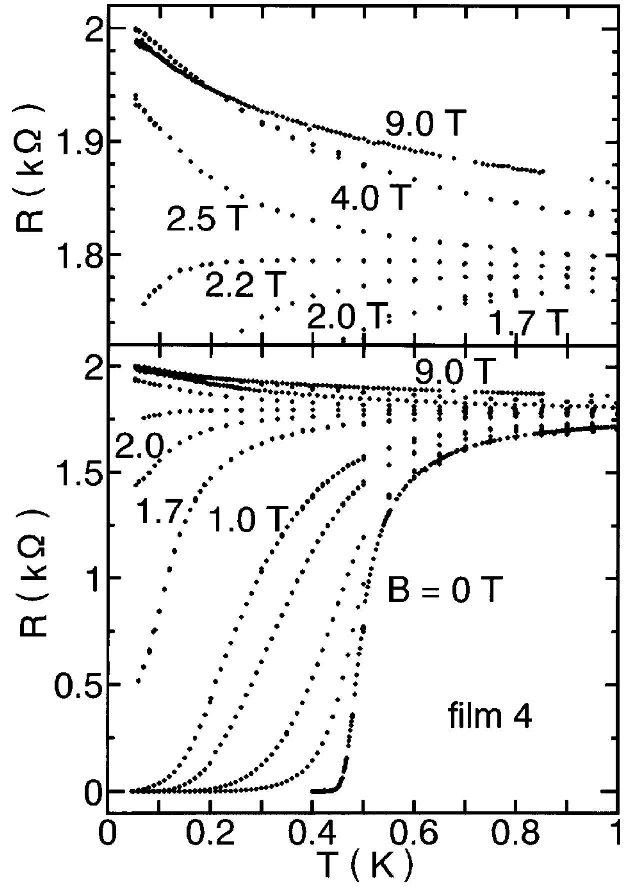

FIG. 2. $R(T)$ of film 4 in various fields. The high-field region is enlarged and shown in the upper panel.

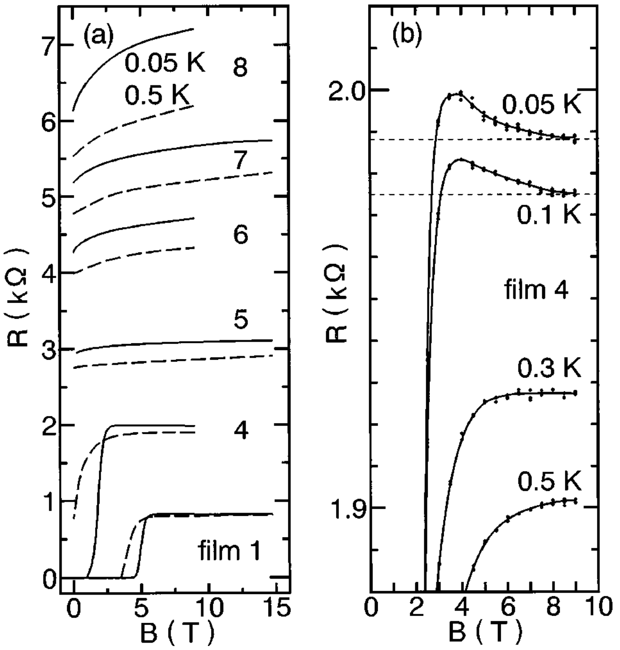

FIG. 3. (a) Magnetoresistance $R(B)$ at $T=0.05$ K (solid lines) and 0.5 K (dashed lines) for a series of films which lie on both sides of the zero-field S-I transition. (b) $R(B)$ of film 4 at different T in fields higher than B_{xxC} (=2.3 T). The anomalous peak is visible at $T=0.05$ and 0.1 K.

$(B \gg B_{xxC})$, the temperature dependence of R becomes weaker. As depicted in the upper panel of Fig. 2, R in the field of 9 T is smaller than that in $B=4$ T below about 0.2 K. Hereafter, we concentrate on the magnetic-field dependence of the resistance (magnetoresistance) $R(B)$ at constant T. Figure 3(a) shows the 0.05-K and 0.5-K isotherms of $R(B)$ for a series of films which lie on both sides of the zero-field S-I transition. First, we examine the insulating films (films 5–8). The positive magnetoresistance is commonly observed irrespective of the temperature: With increasing B, R rises monotonically and shows a sign of saturation at higher fields. The amplitude of the magnetoresistance $\Delta R(B)=R(B)-R(0)$ tends to grow as T decreases and/or the films become more resistive. Plotting G against $\ln B$, we have found that all the data shown in Fig. 3(a) (and the data for film 9) exhibit a $\ln B$ dependence at fields higher than 0.7–1 T; the coefficient of the $\ln B$ term is about $(0.4-0.6)\times e^2/2\pi^2\hbar$ for these films. These results are explained with the 2D weak-localization theory for fermions in the presence of the strong spin-orbit scattering, which probably originates from Mo atoms.

Next, we turn to the superconducting films (films 1–4), where we focus on the field region higher than B_{xxC}. The representative result for film 4 is shown in Fig. 3(b). At high temperatures ($T=0.3$ and 0.5 K), $R(B)$ is a monotonically increasing function of B. However, as the temperature decreases ($T=0.05$ and 0.1 K), a maximum in $R(B)$ occurs at B slightly larger than $B_{xxC}=2.3$ T; with further increasing B, $R(B)$ shows a decrease (i.e., negative magnetoresistance) and tends to a value (shown with a dashed line) that increases with decreasing T. Qualitatively similar behavior of $R(B)$ is observed for other superconducting films.

We consider the possible origins of the negative magnetoresistance. The most simple explanation is that it is resulting from the competition between the positive magnetoresistance due to the superconducting fluctuation and the negative magnetoresistance due to the Anderson localization: At high temperatures, the magnetoresistance is dominated by the superconducting fluctuation. Upon cooling, the superconducting fluctuation contributing to the conductance decreases and at low enough temperatures, $R(B)$ is dominated by quantum fluctuations. In this picture, the decrease in $R(B)$ after the maximum is due to the delocalization effect of the unpaired electrons (fermions) by the magnetic field. However, this picture is not consistent with the result for insulating films (films 5–9) described above, where the magnetoresistance is always *positive* and it is well explained within the framework of the weak-localization theory taking account of the strong spin-orbit interaction. Since the superconducting films (films 1–4) also lie in the weakly localized regime (for fermions), and the strong spin-orbit scattering is certainly present due to higher Mo concentration than in the insulating films, it is natural to expect that the magnetoresistance originating from unpaired electrons should be positive.

Therefore, we consider another possible explanation for the negative magnetoresistance: We seek the origin in (localized) Cooper pairs that are present even at $B>B_{xxC}$. This idea is based on the assumption that B_{xxC} is smaller than an upper critical B_{C2} or B_{xyC}, at which Cooper pairs disappear. Here B_{xyC} $(>B_{xxC})$ is a second critical field determined from the Hall resistance R_{xy}, which was reported by Paalanen, Hebard, and Ruel (PHR) in amorphous InO_x films[8] and later by us in In films.[12] In the absence of the theory describing $R(B)$ for the ‘‘dirty boson’’ system, there is no strong justification for the idea proposed here. Moreover, on the experimental side, the determination of B_{C2} is generally difficult. We attempted to measure R_{xy} to obtain B_{xyC}, however, R_{xy} ($\sim$0.01–0.1 Ω) was too small to accurately determine B_{xyC}. Thus we tentatively regard a characteristic field $B^*(T)$ above which the negative magnetoresistance is no longer visible as $B_{C2}(T)$: the values of $B^*(0.05\text{ K})$ thus estimated are $\sim$15 and $\sim$10 T for films 1 and 4, respectively, which are much larger than $B_{xxC}(T\rightarrow 0)=5.5$ T (film 1) and 2.3 T (film 4). With decreasing temperature from 0.1 to 0.05 K, the amplitude of the peak in $R(B)$ becomes more pronounced, while B^* decreases only slightly or stays almost unchanged, suggesting that the *unusual* field region $[B_{xxC} < B < B^*(T)]$ persists down to $T\rightarrow 0$. If this picture described here is acceptable and the present system is regarded as an ideally homogeneous 2D system, it is not unreasonable to interpret this unusual field region as signaling the $T=0$ Bose-glass insulator. Theoretically,[1] the Bose-insulator phase originates from the strong quantum fluctuations in the phase of the order parameter in 2D at $T=0$. This is consistent with the present result that the unusual field region is observable only at low enough temperatures.

From the present data of $R(B)$, we are not able to obtain any clues suggesting the presence of Cooper pairs on the insulating side of the zero-field S-I transition. This fact, together with finding that the critical resistances for both the zero-field and field-driven S-I transitions are nonuniversal, seems to contradict the ‘‘dirty boson’’ model of Fisher *et al.*. However, we are currently not certain whether these results immediately imply breakdown of this model, because the measurements have been performed at finite temperatures. At finite T (or B), the measured conductance includes the contribution of unpaired electrons (fermions) as well as that of Cooper pairs (bosons).[3,14] When the unpaired electrons are only weakly localized, their contribution to the conductance cannot be negligible even at $T=0.05$ K. Thus, even though the subtle anomaly of $R(B)$ originating from bosons would occur in insulating films at $B>B_{xxC}$ $(=0)$, it might be hidden by the relatively large magnetoresistance arising from unpaired electrons. In addition, at $T>0$ the contribution of unpaired electrons to the conductance will always result in the smaller value of the critical resistance than that for Cooper pairs.[21]

We now comment on the morphology of our films, since it is important to interpret the experimental data properly.[13] Although we have confirmed from the TEM observation that the films used in this study are homogeneous, it is difficult to exclude definitely the possibility for the existence of very small grains, such as Mo grains, whose sizes are below our experimental resolutions. Pure Mo metal is a superconductor with a superconducting transition temperature about 0.9 K. Thus there remains the possibility that the anomalous behavior of $R(B)$ observed at low temperatures may be related to the destruction of local superconductivity within each grain. Nevertheless, we believe such a possibility to be small due to the following reasons: (1) For all of the films studied, there is no sign of local superconductivity in $R(T)$, which is often visible as a sharp drop of $R(T)$ at certain temperatures very close to the bulk transition temperature for clearly granular superconductors: (2) The anomalous magnetoresistance is observed only in superconducting films but not in insulating films. If such grains of Mo might exist in superconducting films, they should also exist and possibly be detected in insulating films.

A nonmonotonic change in $R(B)$ was also found in several 2D superconductors. In granular films of Sn (Ref. 5) and $\mathrm{DyBa_2Cu_3O_{7-x}}$ (Ref. 10), for example, an anomalous peak in the magnetoresistance was reported at low temperatures; however, the behavior is rather different from the present result. An essential difference is that the peak is clearly observed in insulating films. We consider that the most important example to be compared with the present results is that for a-InO_x films by PHR.[8] In this homogeneous system, a maximum in $R(B)$ at $B>B_{xxC}$ and a subsequent decrease in R with increasing B were observed at low temperatures for a superconducting film, though results for insulating films were not reported. Combined with the remarkable finding of B_{xyC} $(>B_{xxC})$, they explained the peak in $R(B)$ based on the notion that an insulator with localized Cooper pairs should have a higher resistance than an insulator with localized single electrons.[8] We notice from Fig. 4 in Ref. 8 that the peak in $R(B)$ occurs at fields slightly higher than B_{xyC}. This implies that the region where the negative magnetoresistance is visible corresponds to the Fermi-insulator phase, leading to the conclusion that the negative magnetoresistance has nothing to do with localized Cooper pairs. This statement is somewhat different from what is discussed in this paper; i.e., the negative magnetoresistance follows from localized Cooper pairs. The disagreement in the interpretation of the negative magnetoresistance, that arises from the discrepancy between B_{xyC} and B^* in Ref. 8, awaits for further investigations. This may not be so serious, if the boundary where localized Cooper pairs disappear is a region of crossover rather than a phase boundary,[8] or $B^*(T)$ approaches B_{xyC} in the limit of $T\rightarrow 0$.

In summary, we have performed systematic studies for both the zero-field and field-driven S-I transitions using a series of ultrathin films of a-$\mathrm{Mo}_x\mathrm{Si}_{1-x}$. For superconducting films, we have observed an anomalous peak in the magnetoresistance and a subsequent decrease in $R(B)$ with increasing B in fields higher than the critical field B_{xxC} at low enough temperatures. In contrast, the magnetoresistance for insulating films is always monotonic and positive irrespective of the temperature, consistent with the 2D weak-localization theory for fermions in the presence of strong spin-orbit interaction. From these results, we suggest a possibility that localized Cooper pairs may exist even on the insulating side of the field-driven S-I transition $(B>B_{xxC})$ at $T\rightarrow 0$.

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[1] M. P. A. Fisher, Phys. Rev. Lett. **65**, 923 (1990).

[2] A. Gold, Z. Phys. B **83**, 429 (1991).

[3] R. Ikeda, Int. J. Mod. Phys. B **10**, 601 (1996).

[4] D. B. Haviland, Y. Liu, and A. M. Goldman, Phys. Rev. Lett. **62**, 2180 (1989).

[5] S. Kobayashi, Physica B (Amsterdam) **152**, 2236 (1988).

[6] A. F. Hebard and M. A. Paalanen, Phys. Rev. Lett. **65**, 927 (1990).

[7] N. Nishida, S. Okuma, and A. Asamitsu, Physica B (Amsterdam) **169**, 487 (1991).

[8] M. A. Paalanen, A. F. Hebard, and R. R. Ruel, Phys. Rev. Lett. **69**, 1604 (1992).

[9] G. T. Seidler and T. F. Rosenbaum, Phys. Rev. B **45**, 10 162 (1992).

[10] K. M. Beauchamp, G. C. Spalding, T. Wang, and A. M. Goldman, Physica B (Amsterdam) **194-196**, 2321 (1994); T. Wang *et al.*, Phys. Rev. B **47**, 11 619 (1993).

[11] J. M. Valles, Jr. *et al.*, Physica B (Amsterdam) **197**, 522 (1994).

[12] S. Okuma and N. Kokubo, Phys. Rev. B **51**, 15 415 (1995); *ibid.* **56**, 410 (1997).

[13] A. M. Goldman and Y. Liu, Physica D **83**, 163 (1995), and references therein.

[14] A. Yazdani and A. Kapitulnik, Phys. Rev. Lett. **74**, 3037 (1995).

[15] S. Okuma, T. Terashima, and N. Kokubo, in *Proceedings of the 9th International Symposium on Superconductivity, Sapporo, 1996*, edited by S. Nakajima and M. Murakami (Springer-Verlag, Tokyo, 1997), p. 191; Solid State Commun. **106**, 529 (1998).

[16] K. Kagawa, K. Inagaki, and S. Tanda, Phys. Rev. B **53**, R2979 (1996).

[17] D. Ephron, A. Yazdani, A. Kapitulnik, and M. R. Beasley, Phys. Rev. Lett. **76**, 1529 (1996).

[18] M. Watanabe, H. Shimada, S. Kobayashi, and Y. Ootuka, J. Phys. Soc. Jpn. **66**, 1419 (1997).

[19] G. Martinez-Arizala *et al.*, Phys. Rev. Lett. **78**, 1130 (1997).

[20] W. L. Johnson *et al.*, J. Appl. Phys. **50**, 4240 (1979).

[21] As shown in Fig. 1(b), the weaker temperature dependence of $G(T)$ in $B=0$ than that in $B=9$ or 14.8 T observed for resistive films (films 6–9) may reflect the presence of Cooper pairs.